

Parameter Analysis of CO₂ Capture with Anti-Sublimation Process

Hua Tian¹, Ke Kang¹, Lingfeng Shi^{2,*} and Rui Sun¹

¹State Key Laboratory of Engines, Tianjin University, Tianjin, 300072, China

²Department of Thermal Science and Energy Engineering, University of Science and Technology of China, Hefei, 230027, China

*Corresponding Author: Lingfeng Shi. Email: slf@ustc.edu.cn

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Abstract: The anti-sublimation CO₂ capture technology has attracted the attention of researchers due to its advantages such as no pollution and high product purity. The anti-sublimation process is the core link of this technology, so the study of this process is of great significance to the low-temperature capture system. At present, there are few research works on the CO₂ anti-sublimation process. In order to study the influence of key parameters on the capture performance during CO₂ anti-sublimation, a one-dimensional steady-state of CO₂ anti-sublimation process in a double pipe heat exchanger was established based on the mixture gas of N₂ and CO₂. The effects of cooling nitrogen inlet temperature, mixture gas velocity and pressure on the CO₂ volume fraction, deposition rate distribution, capture rate and valid capture length are investigated. Optimal operation parameters are obtained, which could be used to provide guidance for parameter setting and design of anti-sublimation heat exchanger.

Keywords: CO₂ capture; anti-sublimation process; parameter analysis

1 Introduction

According to statistics from the International Energy Agency, the global energy-related carbon dioxide emissions in 2018 reached the highest level in history of 3.31 billion tons [1]. Currently, there are four ways for the post-combustion method of CO₂ capture: solvent absorption, solid sorbent adsorption, low-temperature separation, and membrane separation [2]. The anti-sublimation CO₂ capture technology has attracted the attention of researchers due to its advantages such as no pollution, high product purity and so on. In the low-temperature separation and capture system, CO₂ change from gaseous state to solid state directly in the anti-sublimation heat exchanger. Due to the significance of the CO₂ anti-sublimation process in low-temperature CO₂ capture system, it is worth investigating the contributing factors in this process.

Currently, the research on the CO₂ anti-sublimation process mainly focuses on simulation study. Chang et al. [3,4] proposed a heat and mass transfer model for CO₂ anti-sublimation process of landfill gas (CH₄ and CO₂), and showed the effects of mixture gas at different CO₂ mole fractions or N₂ to CO₂ mass flow rate ratios on the performance parameters. Yu et al. [5] developed a numerical model of the heat and mass transfer processes in a CO₂-N₂ gas mixture anti-sublimation cross-flow finned duct heat exchanger system to predict the heat transfer from gas mixture to liquid nitrogen and the anti-sublimation rate of CO₂ in the



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gas mixture. Naletov et al. [6,7] proposed a quasi-nonstationary mathematical model of the anti-sublimation of carbon dioxide from purified flue gases of heat power systems and obtained the optimal operation estimates and engineering solution. Wang et al. [8] developed a transient model for analyzing the CO₂ anti-sublimating in mixture gas, and drew a conclusion that a low energy consumption will be obtained at high concentration and low flow velocity of CO₂ supply.

From the above, these studies mainly focused on the temperature distribution of the cold and hot fluids and the CO₂ deposition rate when the state parameters of the hot and cold fluid inlets (such as the initial CO₂ mole fraction, the cooling nitrogen inlet temperature, etc.) change during the anti-sublimation capture process. In addition, in the study of the carbon dioxide capture system, some researchers have investigated the effect of the mixture gas inlet pressure on capture system performance. Result show that the change of inlet pressure of the mixture gas could have a great impact on the capture system performance, such as Song et al. [9]. The studies about parameter characteristics are summarized in Tab. 1. In terms of operating parameters, there is a lack of the influence of mixture gas pressure on the anti-sublimation process. Besides, in terms of performance parameters, there is a lack of comprehensive consideration of the valid capture length and capture rate, which could guide the optimal design of the heat exchanger equipment in the anti-sublimation process. Based on this point, future research was completed in this article.

Table 1: Research status of CO₂ anti-sublimation process parameters

Author	Operating parameters	Performance parameters
Chang et al. [3,4]	Inlet CO ₂ fraction, Proportion of each component in the mixture gas	Gas and heat exchanger tube wall temperature, Initial anti-sublimation point location, Tube length, CO ₂ deposition rate
Yu et al. [5]	Cooling nitrogen inlet temperature, Number of heat exchanger fins	CO ₂ mass flow rate, deposition rate, deposition thickness
Naletov et al. [6,7]	Time	Gas temperature, CO ₂ mass flow rate, Deposition rate
Wang et al. [8]	Time, Initial wall temperature, Inlet CO ₂ fraction, Flow velocity of mixture gas	Gas temperature, Wall temperature, CO ₂ deposition rate, Capture rate, Energy consumption

In this paper, a one-dimensional steady-state anti-sublimation process numerical model of CO₂ in N₂ and CO₂ mixture gas was developed. Based on a double-pipe heat exchanger, the model considered the influence of the CO₂ deposition on the heat and mass transfer process, and added the valid capture length to performance parameters to evaluate the capture result comprehensively. Study on the effects of cooling nitrogen inlet temperature, mixture gas flow rate and pressure change on CO₂ volume fraction, deposition rate and distribution, initial anti-sublimation point location, valid capture length and capture rate has been performed. This can not only determine the appropriate boundary conditions of the anti-sublimation process, but also provide guidance for the design of heat exchanger length and position of requiring heat transfer enhancement.

2 Numerical Model

As shown in Fig. 1, a one-dimensional steady-state model of CO₂ anti-sublimation process is proposed based on a double-pipe heat exchanger. The mixture gas and cooling nitrogen flow reversely in the heat exchanger. The subscript m represents the mixture gas, and the subscript c means the cooling nitrogen, and the temperature of the tube near the mixture gas and cooling nitrogen are $T_{w,m}$ and $T_{w,c}$ respectively.

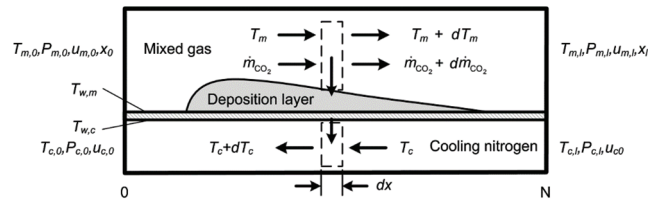


Figure 1: Model of double-pipe heat exchanger

The entire length of heat exchanger is divided into N nodes. When the temperature of the mixture gas drops to the anti-sublimation temperature corresponding to the CO₂ partial pressure at that node, CO₂ anti-sublimation process occurs. The main assumptions are as follows:

- (1) The parameters have no radial gradient and it is a one-dimensional steady-state model.
- (2) The pressure drop, axial heat conduction and radiant heat leakage are ignored.
- (3) The mixture gas and cooling nitrogen are considered to be an ideal gas.

2.1 CO₂ Anti-Sublimation Temperature

CO₂ anti-sublimation temperature is calculated by PR equation of state and gas-solid fugacity equation [10].

The solid phase CO₂ product obtained by the CO₂ anti-sublimation capture technology has high purity and it can reach 99% without further purification [7,11]. Therefore, it can be considered that the deposition layer is composed of pure solid CO₂. In order to calculate the frosting temperature of CO₂ in different components and different pressures in the binary system, the gas-solid fugacity equation is established, as shown in the following equation:

$$\Delta f_{CO_2}^{V-S}(T_m) = x_{CO_2} \phi_{CO_2}^V P_m - P_{CO_2Solid}^{Sat} \phi_{CO_2}^{Sat} \times \exp \left[v_{CO_2Solid} (P_m - P_{CO_2Solid}^{Sat}) / RT_m \right] = 0 \quad (1)$$

where $\Delta f_{CO_2}^{V-S}(T_m)$ is the gas-solid phase fugacity difference at T_m , x_{CO_2} is the mole fraction of CO₂ in vapor phase, $\phi_{CO_2}^V$ is the CO₂ vapor phase fugacity, P_m and T_m are the mixture gas pressure (kPa) and temperature (K) respectively, $P_{CO_2Solid}^{Sat}$ is the CO₂ saturation pressure corresponding to the mixture gas temperature, $\phi_{CO_2}^{Sat}$ is the mixture gas temperature corresponding to the fugacity coefficient of pure CO₂, v_{CO_2Solid} is the molar volume of solid CO₂.

The saturated CO₂ pressure can be obtained by the following equation [12]:

$$\ln \left(\frac{P_{CO_2Solid}^{Sat}}{P_{tr}} \right) = \frac{T_{tr}}{T} \times \left[a_1 \left(1 - \frac{T}{T_{tr}} \right) + a_2 \left(1 - \frac{T}{T_{tr}} \right)^{1.9} + a_3 \left(1 - \frac{T}{T_{tr}} \right)^{2.9} \right] \quad (2)$$

where $T_{tr} = 216.592$ K, $P_{tr} = 0.51795$ MPa, $a_1 = -14.740846$, $a_2 = 2.4327015$, $a_3 = -5.3061778$.

2.2 Anti-Sublimation Process Model

The sum of convective heat from gas mixture and CO₂ latent heat equal to the convective cooling by the cooling nitrogen, and equal to the heat transfer of the wall:

$$h_m P_1 (T_m - T_{w,m}) + i_{ig} \left(- \frac{d\dot{m}_{CO_2}}{dx} \right) = \frac{2\pi\lambda_d (T_{w,m} - T_{w,c})}{\ln \frac{r_2 + th}{r_1}} = h_c P_2 (T_{w,c} - T_c) \quad (3)$$

P_1 and P_2 are the heat transfer perimeters (m) on both sides of the inner tube, i_{ig} is the latent heat of phase transition of CO_2 (J/kg), $\Delta\dot{m}_{\text{CO}_2}$ is CO_2 deposition rate ($10^{-6} \text{ kg}\cdot\text{m}^{-1}\cdot\text{s}^{-1}$), λ_d is the equivalent thermal conductivity of the wall and the deposition layer ($\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$), r_1 and r_2 are the inner and outer radius of the inner tube (m), and h is the heat transfer coefficient ($\text{W}\cdot\text{m}^{-2}\cdot\text{K}^{-1}$).

According to the equal heat flux, the equivalent thermal conductivity λ_d can be obtained by the equation:

$$\frac{2\pi dx(T_m - T_c)}{\frac{\ln\left(\frac{r_2 + th}{r_2}\right)}{\lambda_d}} = \frac{2\pi dx(T_m - T_c)}{\frac{\ln\left(\frac{r_2}{r_1}\right)}{\lambda_w} + \frac{\ln\left(\frac{r_2 + th}{r_2}\right)}{\lambda_s}} \quad (4)$$

where th is the thickness of the deposition layer (m). Assuming that the deposition layer is evenly distributed in the radial and circumferential directions, then:

$$th = \sqrt{\frac{dV_{\text{CO}_2}}{\pi dx} + r_2^2} - r_2 \quad (5)$$

where dV_{CO_2} is the CO_2 deposition volume (m^3) on the each node, $dV_{\text{CO}_2} = \pi(R_2^2 - r_2^2)dx$, R_2 is the equivalent outer radius (m) of the tube wall and the deposition layer.

The density of the solid CO_2 (kg/m^3) is [13]:

$$\rho_s = -0.004T^2 + 0.1T + 1679.8 \quad (6)$$

The steady-state flow energy balance equations of mixture gas and cooling nitrogen are as follows:

$$(\dot{m}_{\text{N}_2}c_{p,\text{N}_2} + \dot{m}_{\text{CO}_2}c_{p,\text{CO}_2})\frac{dT_m}{dx} = -h_mP_1(T_m - T_{w,m}) \quad (7)$$

$$\dot{m}_c c_{p,c} \frac{dT_c}{dx} = h_c P_2 (T_c - T_{w,c}) \quad (8)$$

where \dot{m} is the mass flow rate (kg/s), c_p is the constant pressure specific heat capacity ($\text{J}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$).

$$h_c = \frac{\lambda_c Nu_c}{d_1} \quad (9)$$

$$Nu_c = 0.023 Re_c^{0.8} Pr_c^{0.4} \quad (10)$$

The mass flow rate of N_2 in the mixture gas is unchanged. Before the mixture gas cools down to the CO_2 anti-sublimation temperature, the CO_2 mass flow rate remains unchanged; after reaching the anti-sublimation temperature, the CO_2 mass flow rate decreases.

$$-\frac{d\dot{m}_{\text{N}_2}}{dx} = 0 \quad (11)$$

$$-\frac{d\dot{m}_{\text{CO}_2}}{dx} = \begin{cases} 0 & , x < x_0 \\ h_D P_1 [\rho_{\text{CO}_2} - \rho_{\text{Sat}(T_{w,m})}] & , x \geq x_0 \end{cases} \quad (12)$$

where h_D is the mass transfer coefficient (m/s), ρ_{CO_2} is the CO_2 density (kg/m^3), $\rho_{\text{Sat}(T_{w,m})}$ is the CO_2 saturation density (kg/m^3) at the wall temperature.

The mass flow rate and density of N₂ and CO₂ in the mixture gas are expressed as:

$$\dot{m}_{N_2} = \rho_{N_2} \bar{u}_m A_m, \quad \rho_{N_2} = \frac{P_m - P_{CO_2}}{R_{N_2} T_m} \quad (13)$$

$$\dot{m}_{CO_2} = \rho_{CO_2} \bar{u}_m A_m, \quad \rho_{CO_2} = \frac{P_{CO_2}}{R_{CO_2} T_m} \quad (14)$$

where \bar{u}_m is the average flow rate of the mixture gas (m/s), A_m is the mixture gas flow area (m²), P_{CO_2} is the partial pressure of CO₂ in the mixture gas (kPa), R_{N_2} , R_{CO_2} are the gas constant of N₂, CO₂ (J·kg⁻¹·K⁻¹).

The heat and mass transfer coefficient can be calculated by analogy with fully developed flow heat and mass transfer:

$$\frac{h_m}{(\rho_{N_2} + \rho_{CO_2}) c_{p,m} \bar{u}_m} Pr^{2/3} = \frac{h_D}{\bar{u}_m} Sc^{2/3} = \frac{f}{2} \quad (15)$$

The friction coefficient f is a function of Reynolds number Re :

$$f = \begin{cases} \frac{16}{Re} & (Re < 2300) \\ \frac{0.046}{Re^{0.2}} & (Re \geq 2300) \end{cases} \quad (16)$$

The Schmidt number Sc is related to the diffusion coefficient D (m²/s):

$$Sc = \frac{\nu}{D} = \frac{\mu}{\rho D} \quad (17)$$

where ν , μ are the kinematic viscosity coefficient (m²/s), the momentum viscosity coefficient (m²/s).

The gas diffusion coefficient in N₂-CO₂ binary system as [6]:

$$D_{N_2-CO_2} = D_0 \left(\frac{T}{T_0}\right)^m \frac{P_0}{P} \quad (18)$$

where $m = 3/2$, $D_0 = 0.0000166567$ m²/s, $T_0 = 298$ K, $P_0 = 100$ kPa.

The calculation of CO₂ anti-sublimation process is shown in Fig. 2. The main input parameters are the gas inlet pressure, temperature, flow rate, mixture gas inlet CO₂ volume fraction and the initial iteration value of cooling nitrogen outlet temperature. Finally get parameters such as CO₂ deposition rate, volume fraction, and capture rate.

2.3 Performance Indicator

In this paper, there are four indicators to evaluate the performance of the anti-sublimation capture process:

(1) Capture rate: the ratio of CO₂ anti-sublimation amount on the total amount of CO₂ in the inlet gas mixture.

$$\text{Capture rate} = \frac{\Delta m_{CO_2}}{m_{CO_2}} \quad (19)$$

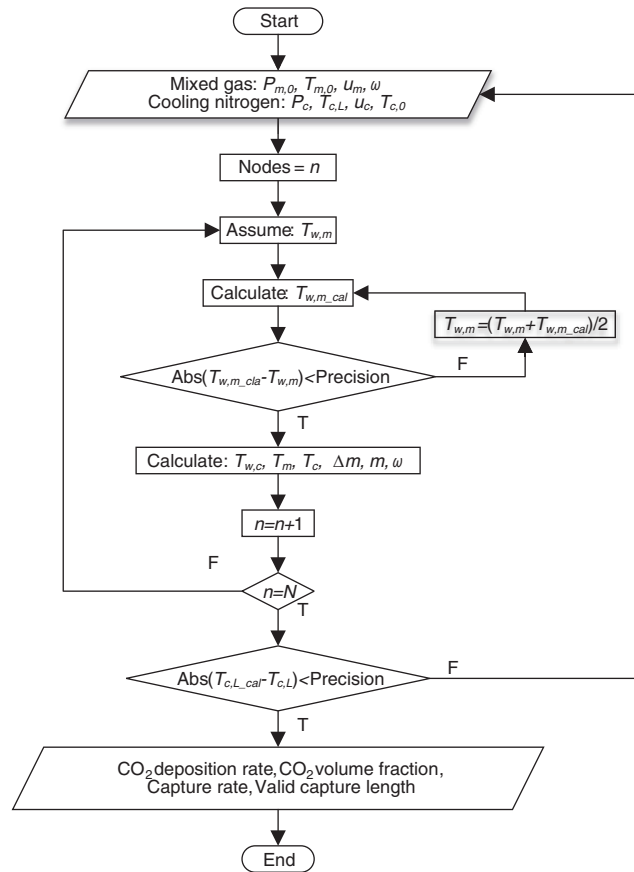


Figure 2: Flow chart of procedural programming in CO₂ anti-sublimation

(2) Initial anti-sublimation point position x_1 : the point where CO₂ begins changing from the gas phase to the solid phase, as shown in Fig. 3a. The earlier the initial anti-sublimation point is, the more uniform heat transfer between two fluids.

(3) Valid capture length L_1 : as shown in Fig. 3a, it is derived from the CO₂ separation target proposed by the US Department of Energy (95% purity and above 90% separation rate) [14]. Considering that the purity of the CO₂ product obtained by the low-temperature capture technology can generally reach 99%, the distance from the mixture gas inlet to the position of 90% capture rate can be defined as the valid capture

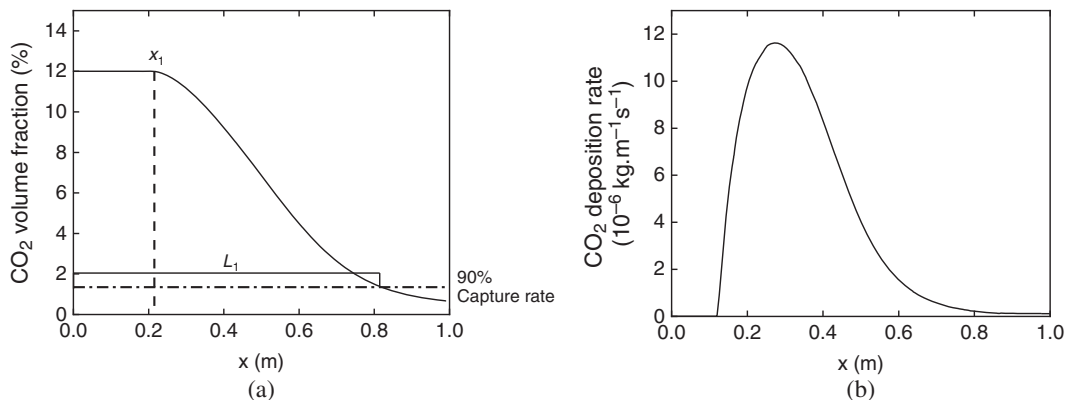


Figure 3: Initial anti-sublimation point, valid capture length and CO₂ anti-sublimation rate

length. The longer the valid capture length, the more uniform the anti-sublimation distribution of solid CO₂, and the smaller the impact on the heat transfer heterogeneity during the anti-sublimation process.

(4) CO₂ anti-sublimation rate distribution: as shown in Fig. 3b, the physical meaning of the anti-sublimation rate is the mass of CO₂ anti-sublimation per unit time and tube length, and its distribution trend represents the distribution of solid CO₂ deposition. The lower CO₂ peak deposition rate and uniform distribution can increase the capture operation time, and reduce the excessive local thermal resistance.

3 Simulation Results

In this model, tube length $L = 1$ m, the number of nodes $N = 200$, the inner and outer radius of the inner tube are $r_1 = 2$ mm, $r_2 = 3$ mm, and the inner radius of outer tube is $r_3 = 4$ mm. At the inlet of gas mixture, $T_{mix}(0)$ is fixed at 298.15 K, the inlet CO₂ volume fraction is given at 12%, the cooling nitrogen inlet pressure is 200 kPa, and its flow rate is 3.5 m/s.

3.1 Influence of Cooling Nitrogen Inlet Temperature

In order to analyze the individual effect of cooling nitrogen inlet temperature on the capture performance, only this parameter is changed while other parameters are kept constant. The inlet pressure of the mixture gas is 200 kPa and its flow velocity is 0.3 m/s. The inlet temperature of cooling nitrogen varies between 130 K and 160 K, and its influence on the anti-sublimation process is shown in Figs. 4 and 5.

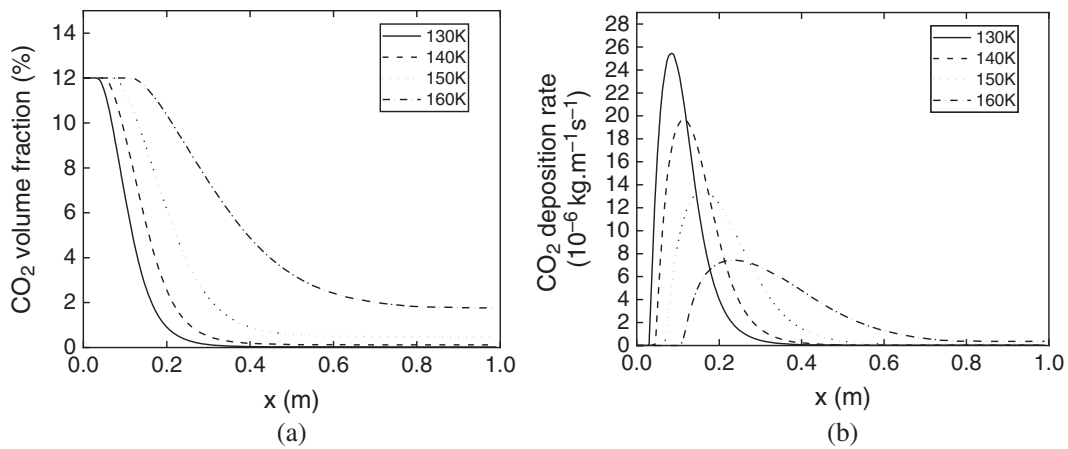


Figure 4: Effects of coolant inlet temperature on concentration and deposition rate of CO₂

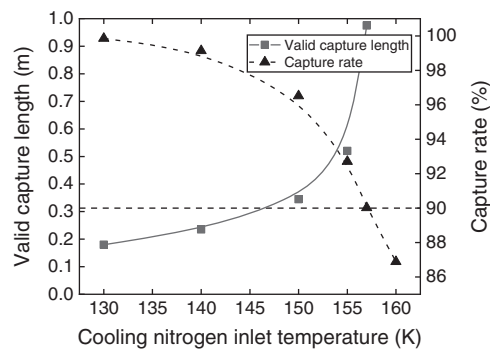


Figure 5: Effects of coolant inlet temperature on valid capture length and capture rate

As shown in Fig. 4a, when the inlet temperature of the cooling nitrogen increases from 130 K to 160 K, the initial anti-sublimation point position moves from 0.03 m to 0.12 m, meanwhile, the volume fraction of CO₂ changes smoothly, and the mixture gas outlet CO₂ volume fraction increases from 0.02% to 1.76%.

In Fig. 4b, The CO₂ deposition rate decreases significantly, and its peak value decreases from $25.73 \cdot 10^{-6} \text{ kg}\cdot\text{m}^{-1}\cdot\text{s}^{-1}$ to $7.45 \cdot 10^{-6} \text{ kg}\cdot\text{m}^{-1}\cdot\text{s}^{-1}$ when the inlet temperature of cooling nitrogen increases. Meanwhile, the deposition rate distribution becomes smooth. When the cooling nitrogen inlet temperature is 130 K, the deposition rate distribution is concentrated between 0.03 and 0.34 m. While the deposition rate distribution corresponding to 160 K is concentrated between 0.11 m and 0.73 m. The uniform distribution of solid CO₂ deposition is good for heat and mass transfer process and effectively utilize the entire length of heat transfer.

As shown in Fig. 5, when the inlet temperature of cooling nitrogen rises from 130 K to 160 K, the valid capture length increases from 0.18 m to 0.99 m, so the problem of excessive local deposition was reduced. When the inlet temperature of the cooling nitrogen rises to 153 K, the valid capture length changes significantly. However, the capture rate decreases rapidly, from 99.85% to 86.92%. When the minimum capture requirement is 90%, the corresponding cooling nitrogen inlet temperature is about 157.5 K. It can be seen that the inlet temperature of cooling nitrogen has a great influence on the valid capture length and capture rate. In the experimental operation, the cooling nitrogen inlet temperature should be set by comprehensively considering the anti-sublimation performance parameters.

3.2 Influence of Mixture Gas Flow Velocity

Calculation data in Figs. 6 and 7 are obtained under following conditions: the inlet temperature of cooling nitrogen is 150 K, the inlet pressure of mixture gas is 200 kPa, and its flow velocity varies from 0.2 m/s to 0.5 m/s.

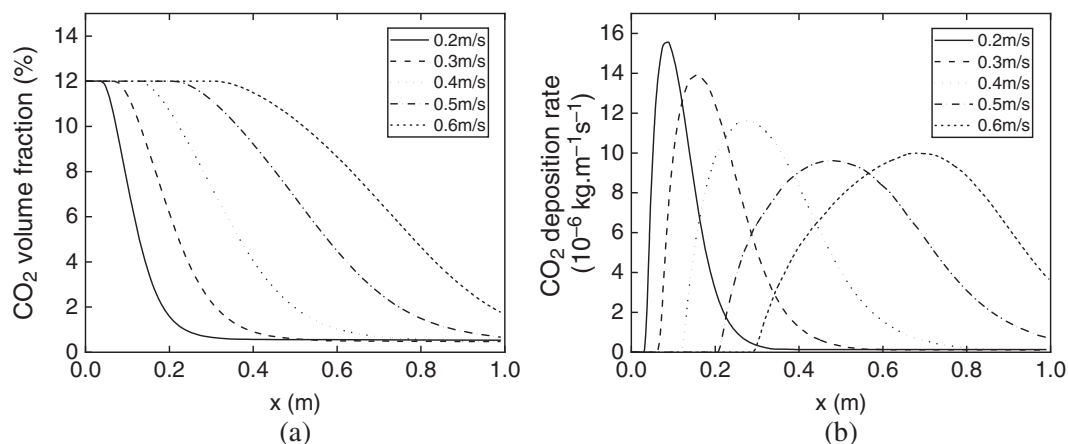


Figure 6: Effects of mixture gas velocity on concentration and deposition rate of CO₂

As shown in Fig. 6a, when the flow rate of mixture gas rises from 0.2 m/s to 0.6 m/s, the position of the initial sublimation point moves from 0.04 m to 0.365 m, and the change of CO₂ volume fraction becomes gentle. In Fig. 6b, when the mixture gas flow rate is 0.5 m/s, the peak deposition rate is the lowest, and the distribution of the deposition rate is smooth. When the mixture gas flow rate is 0.2 m/s, the deposition rate distribution is concentrated between 0.03 and 0.34 m, and the corresponding distribution is concentrated between 0.21 and 0.99 m when the flow rate ascends to 0.5 m/s, which will extend the capture operating time.

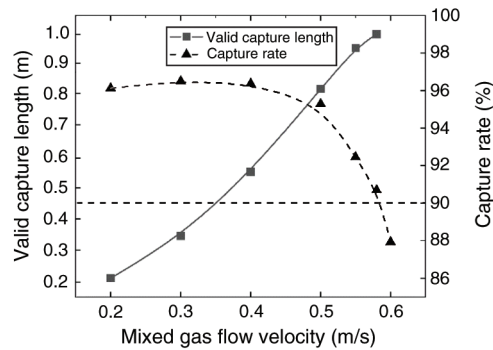


Figure 7: Effects of mixture gas velocity on valid capture length and capture rate

As illustrated in Fig. 7, with the mixture gas flow rate increases from 0.2 m/s to 0.6 m/s, the valid capture length increases significantly, from 0.21 m to 0.96 m, and the capture rate drop from 96.13% to 87.9%. When the mixture gas flow rate exceeds 0.5 m/s, as the flow rate of the mixture gas increases, the capture rate drops significantly. When the mixture gas flow rate further increases to 0.58 m/s, the capture rate falls to 90%. It can be concluded that the mixture gas velocity has a great impact on valid capture length and capture rate, and get a better performance in both sides when the velocity is 0.5 m/s.

3.3 Influence of Mixture Gas Pressure

Figs. 8 and 9 show the influence of mixture gas pressure on the capture performance when the mixture gas pressure varies between 100 kPa and 400 kPa and its velocity is 0.3 m/s. The inlet temperature of cooling nitrogen is 150 K and its pressure is 200 kPa.

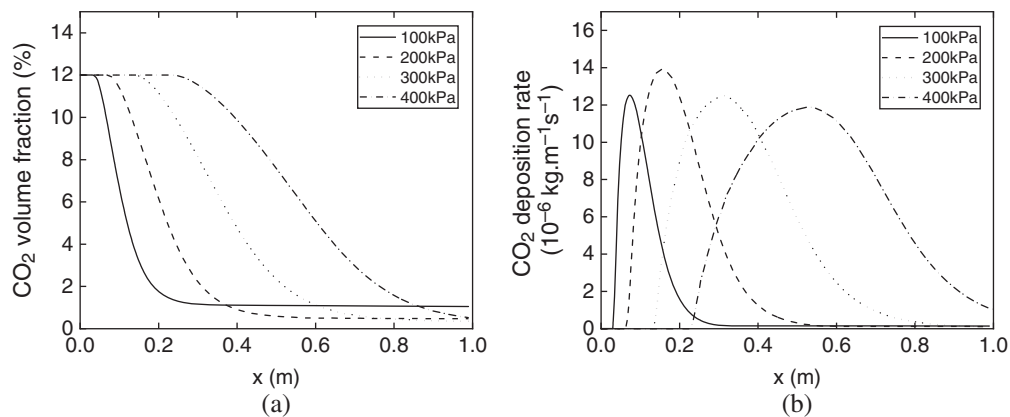


Figure 8: Effects of mixture gas pressure on concentration and deposition rate of CO₂

As depicted in Fig. 8a, with the mixture gas pressure increases from 100 kPa to 400 kPa, the initial anti-sublimation point moves from 0.04 m to 0.24 m, and the change of CO₂ volume fraction tended to be gentle. It can be seen that the mixture gas pressure has a great impact on the position of the initial anti-sublimation point and the volume fraction of CO₂. In Fig. 8b, as the mixture gas pressure increases, the peak value of CO₂ deposition rate of each curve was similar, but the deposition rate distribution from 0.03 m~0.31 m to 0.23 m~1 m, the problem of excessive local deposition is significantly reduced.

In Fig. 9, the mixture gas pressure increases from 100 kPa to 400 kPa, and the valid capture length increases from 0.23 m to 0.83 m, indicating that the larger mixture gas pressure is conducive to make

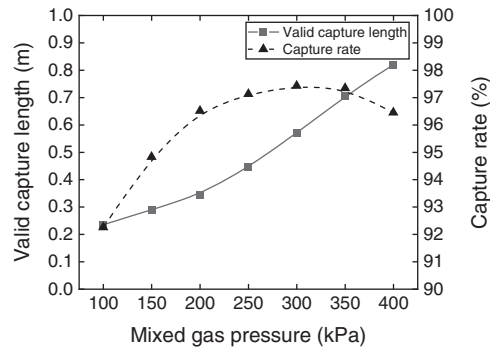


Figure 9: Effects of mixture gas pressure on valid capture length and capture rate

better use of the entire section of heat transfer length. Meanwhile, the capture rate increases slightly from 92.27% to 97.5% and then decreased to 96.4%, indicating that the increment of mixture gas pressure can improve the capture performance, but excessive increment of the mixture gas pressure will lead to the problem of CO₂ capture rate decrease.

3.4 Optimal Parameter Setting Results

With the aim that valid capture length ratio higher than 70% and capture rate higher than 90%, the optimized operating parameters range are shown in Tab. 2:

Table 2: Operation parameters range

Operating parameters	Range
Inlet temperature of cooling nitrogen (K)	155.5~157.5
Flow velocity of mixture gas (m/s)	0.45~0.58
Pressure of mixture gas (kPa)	200~400
Position of strengthen the heat transfer (m)	0.5~0.7

4 Conclusions

In this work, a one-dimensional steady-state model of the CO₂ anti-sublimation process of N₂-CO₂ binary mixture gas was established. Study on the effects of cooling nitrogen inlet temperature, mixture gas flow rate and pressure change on CO₂ volume fraction, deposition rate and distribution, initial anti-sublimation point location, and valid capture length and capture rate has been performed. The conclusions are drawn as follows:

(1) Increasing cooling nitrogen inlet temperature, mixture gas flow rate and pressure can make the position of the initial anti-sublimation point move backward. Meanwhile, the change of CO₂ volume fraction tends becomes smooth, the distribution of anti-sublimation rate tends to be uniform, and the valid capture length increases. But the CO₂ capture rate drops significantly when the inlet temperature of cooling nitrogen increases. There exists an optimal mixture gas flow rate and pressure for achieving the maximum CO₂ capture rate.

(2) With the aim that valid capture length ratio higher than 70% and 90% capture rate, the suggested operating parameters are as follows: the inlet temperature of the cooling nitrogen is between 155.5 and 157.5 K, the flow rate of the mixture gas varies between 0.45 m/s and 0.58 m/s, the pressure of the mixture gas is a variable in range of 200 kPa~400 kPa, and the suggested position of heat transfer

enhancement is between 0.5 m and 0.7 m. Considering that the adjustment of pressure in actual operation often brings additional energy consumption, the cooling nitrogen inlet temperature and mixture gas flow rate should be adjusted first to obtain a better valid capture length.

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Conflicts of Interest: The authors declare that they have no conflicts of interest to report regarding the present study.

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