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A mini-review on cryogenic carbon capture technology by desublimation: theoretical and modeling aspects

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Carbon capture, utilization, and storage (CCUS) technologies are the most effective methods to reduce CO₂ emissions from fossil fuel power plants. Of the different CCUS technologies, cryogenic carbon capture (CCC) methods are the most mature technology as they can obtain remarkably high CO₂ recovery and purity (99.99%). The significant advantage of the CCC process is that it can be easily retrofitted to existing systems and can handle the gas stream's impurities. Different desublimation-based CCC technologies like Cryogenic packed bed, Anti sublimation, External cooling loop, CryoCell process and Novel low-cost CO₂ capture technology (NLCCT) are reported in the literature. The significant limitations of these processes are the continuous removal of the dry ice into storage tanks. For the efficient design of CCC systems, accurate prediction of the phase equilibria data and modeling of the frost formation is called for. This paper reviews the recently reported cryogenic desublimation technologies and analyses the various challenges in making them economically viable. The article also examines the different heat and mass transfer models employed to model CO₂ frost formation.

KEYWORDS

carbon capture, cryogenic technology, desublimation, frost modeling, EOS

1 Introduction

According to the International Energy Agency's Breakthrough Agenda Report 2022 (IEA, 2022), the world is still far from meeting the Paris agreement goal of limiting global temperature rise to 1.5°C this century. The concentration of CO₂ in the atmosphere has reached an all-time high, rising from 280 ppm in the year 1750–420.23 ppm in 2022 (NOAA, 2022). An average annual rise of 2.4 ppm has occurred for the past 10 years. In 2021 global CO₂ emissions reached the highest level ever reported, with the majority coming from the combustion of fossil fuels (IEA, 2022b, 2022). According to the IPCC scenarios, limiting global warming to less than 1.5°C demands a 43% reduction in greenhouse gas emissions by 2030 (IPCC, 2022).

CCUS can lower carbon emissions while encouraging the clean use of conventional fossil fuels. Absorption, Adsorption, Membrane, Hydrate, Microalgae, Chemical looping combustion and cryogenic carbon capture (CCC) are some of the most widely researched CCUS strategies (Song et al., 2018). In recent years CCC has gained wide acceptance as they provide very high CO₂ purity and are devoid of chemical reagents and unwanted pollution (Berstad et al., 2013; Guo et al., 2019; Naquash et al., 2022). CCC makes

TABLE 1 Energy consumption comparison of various desublimation methods.

Process	Mole fraction of CO ₂ in flue gas (%)	CO ₂ capture %	Energy consumption (GJ/t CO ₂)	Cost (\$)	Method	Reference
Cryogenic packed bed	13	99	1.8	55–130	Experiment	Tuinier et al. (2011), Ali (2014), Song et al. (2019)
External cooling loop (ECL)	4	90	0.74		Experiment and simulation	Jensen et al. (2015)
CryoCell technology	20 to 35	34			Experiment and modeling	Hart and Gnanendran (2009)
AnSU process	12	90	1.18		Experiment	Pan et al. (2013)
NLCCT	6.7	99	0.63	7.47/t CO ₂ - 12.64/t CO ₂	Modelling	De et al. (2022)

use of the differences in the condensation (cryogenic distillation techniques) or desublimation points (cryogenic desublimation techniques) of the constituent gases in the flue gas (FG) to capture CO₂. In the distillation techniques, the CO₂ is separated and recovered in liquid form through distillation columns. Although high-purity CO₂ is obtained, they have the disadvantage of being highly energy intensive as the system needs to be maintained at very high pressures to avoid the formation of CO₂ frost (Font-Palma et al., 2021). Distillation methods are energy efficient only for gases with a high concentration of CO₂ (Co) (>50%) (Shen et al., 2022). Stirling cooler (Song et al., 2012), cryogenic distillation (Yousef et al., 2017) etc., are some examples. In the case of desublimation techniques, CCC technology uses the unique thermodynamic properties of CO₂ that at atmospheric pressures and -78.5°C , CO₂ desublimates to a solid state directly from the gaseous state (Baxter et al., 2009; Hart and Gnanendran, 2009). The CO₂ thus obtained (90%–95%) is collected in the form of a solid and can be easily transported and stored. The significant limitations include process blockage due to CO₂ frost and removing the captured frost to storage tanks. Packed bed (Tuinier et al., 2010) and NLCCT (De et al., 2022) are some examples.

For developing the new and efficient design of capturing devices, a detailed understanding of the existing capture technologies and the various modeling strategies is required. The existing technologies should be evaluated based on the ECC, capture % and purity, and the heat and mass transfer models must be analyzed based on their physical significance and prediction capability. This review provides an exhaustive literature survey of the current capture technologies based on the above. The study presents an overview of recent developments in CCC techniques and their challenges. Additionally, the paper assesses the existing approaches for simulating heat and mass transfer during frost formation.

2 CCC by desublimation techniques

Different types of CCC by desublimation techniques have been developed recently: Packed bed, Anti-sublimation, External cooling loop, CryoCell and Novel low-cost CO₂ capture technology (NLCCT). A technical comparison of these methods is provided in Table 1.

2.1 Cryogenic packed bed

CCC by the packed bed is a novel technique. Packed beds made of glass beads or steel beads are used to capture CO₂. The capture process by packed bed involves cooling, capture and regeneration cycles (Ali, 2014). The packed bed is first cooled to cryogenic temperatures and then subjected to FG, where the CO₂ in the gas is desublimated onto the bed as dry ice (Tuinier et al., 2010). Once the packed bed is saturated with dry ice, it must be removed, and the packed bed should be regenerated for the next cycle. Tuinier et al. (2011) proposed a dynamic packed bed (moving bed) method which recovers CO₂ at a rate of 99% and uses 1.8 GJ/t CO₂. Babar et al. (2021) have suggested using multiple packed beds/switched beds. Once the first bed is saturated, the process changes to the other, thus making continuous capture possible. The limitation of this process is the constant removal of the deposited frost from the packed bed and maintaining all the other components frost free (Tuinier et al., 2011; Song et al., 2019).

2.2 External cooling loop (ECL)

Baxter et al. (2009) developed a hybrid CCC system using an ECL. The system makes use of a refrigerant to cool the flue gas. The flue gas is initially dried and cooled to a temperature just above the frost point of CO₂. The CO₂ is then condensed in a desublimating heat exchanger, where it is precipitated in the form of dry ice. The dry ice is then separated and pressurized to a liquid state for storage and transportation. As the heat exchangers are prone to frost, the system components must be maintained at higher pressures, which incurs additional energy consumption for compression. This method provided 99% capture with energy consumption (ECC) of 0.74 GJ/t CO₂ (Baxter et al., 2009; Jensen et al., 2015).

2.3 CryoCell technology

CryoCell technology operates on a similar principle as the ECL method. The dried FG is cooled to a temperature just above the freezing point of CO₂ by a heat exchanger using the liquid CO₂ and treated gas, returning from the separator. The CO₂ is then expanded using a J-T expansion valve to create a three-phase mixture, from

which CO₂ frost is segregated using a three-phase separator. The frosted CO₂ deposited at the bottom of the separator is removed as a liquid by melting the frost with the help of a heater (Hart and Gnanendran, 2009). The liquid CO₂ is pumped to the necessary disposal pressure while the gas is compressed to sales gas criteria. The process is energy efficient only at higher Co (>20%), and the use of compressors increases ECC (Song et al., 2019). For 20%–35% Co, the system produces a capture efficiency of 34% (Hart and Gnanendran, 2009).

2.4 Anti sublimation unit (AnSU)

Clodic et al. (2003) proposed AnSU. This method uses a sequence of expanders and evaporators to capture CO₂ on the low-temperature frost evaporators (LTFE). Two LTFEs are used in this process, one for CO₂ capture and the other for regeneration. A series of heat exchangers cool the FG, and CO₂ gets deposited as frost on the surface of the LTFE (Eide, 2005). For the 12% Co, the process gives an efficiency of 90% with an ECC of 1.18 GJ/t CO₂ (Pan et al., 2013). As the development of the CO₂ frost layer on the heat exchanger surface affects its effectiveness, the heat exchanger materials should have higher thermal conductivity and mechanical stresses (Song et al., 2019).

2.5 Novel low-cost CO₂ capture technology (NLCCT)

De et al. (2022) proposed an NLCCT model that can capture CO₂ with low cost and low water consumption. This method uses cold nitrogen gas refrigerators, regenerative cooling and efficiently constructed cooling chambers to desublimates the CO₂. The challenge involved is the requirement of higher heat transfer rates for the heat exchangers and the design of cooling chamber plates to capture CO₂ effectively. For 6.7% Co, the model shows a capture efficiency of 99% with an ECC of 0.63 GJ/t CO₂.

Advancements in CCC methods are severely limited by the high energy requirements and the capture cost involved (Tuinier et al., 2010; Song et al., 2019). It was observed that ECC rises exponentially as the Co decreases below 10% (Clodic et al., 2005). Additionally, as the CO₂ is collected in the form of frost, an accurate representation of the frost point and valid models that can predict frost growth is called for.

3 Frost formation and its modeling

Frost formation is quite common in cryogenics, aerospace, and refrigeration applications. Frost layers are porous patterns with ice particles and air holes. Frost formation is advantageous in carbon capture applications. However, it can be undesirable in certain others as they can increase resistance to heat transfer and clog flow channels and even cause a system failure (Wu and Webb, 2001; Wu et al., 2007; Ma et al., 2018). In CCC, frost formation occurs when the FG is cooled below the desublimation temperature of CO₂ corresponding to its partial pressure. Sun et al. (2020) have observed that the frost development in cryogenic conditions (<−150°C) is different from frosting under refrigeration conditions (>−20°C)

(Hayashi et al., 1977). The cryogenic frost formation process is divided into three stages: crystal growth, frost layer growth and frost layer full growth phase (Piuco et al., 2008). In the first phase, ice crystal's initial development occurs; in the subsequent two phases, the frost growth with an increase in frost density and thickness occurs (Dave et al., 2017). To predict frost formation and its growth, frost modeling is essential. Literature shows a lack of studies regarding CO₂ frost compared to water vapor frosting.

3.1 Frosting modelling

Frost model development is critical to forecast the frost point and to model frost formation and growth. CO₂ frost point is frequently predicted by Equation of states (EOS), and frosting models are employed to predict frost formation and growth. Frosting models can be generally categorized into empirical correlations and analytical methods (Schneider, 1978; Sommers et al., 2017; Wu et al., 2017).

3.1.1 Thermodynamic modelling by EOS

In CCC methods, CO₂ desublimates from the vapor state and freezes from the liquid state. This makes accurate prediction of the phase equilibrium of CO₂ mixtures essential (de Guido et al., 2014; Nasrifar and Moshfeghian, 2020). In cases where solid CO₂ forms from a vapor or liquid state, describing Solid vapor equilibrium (SVE) and solid-liquid equilibrium (SLE) is essential and in cases where solid CO₂ forms in the presence of liquid and vapor, predicting Solid-Liquid-Vapor (SLVE) is essential (Gu et al., 2018). Liquid and vapor phases are described by cubic equations of states such as Peng-Robinson (PR), Soave-Redlich-Kwong (SRK) and Nasrifar and Bolland (NB) (Soave, 1972; Peng and Robinson, 1976; Nasrifar and Bolland, 2006). Solid CO₂ is efficiently described by the term fugacity (f) (Eq. a), which consists of CO₂ sublimation pressure, vapor fugacity coefficient and Poynting correction factor (Li et al., 2016).

$$f_{\text{pureCO}_2}^s(T, P) = p_{\text{CO}_2}^{\text{sat}} \phi_{\text{CO}_2}^{\text{sat}} \exp \left[\frac{v_{\text{CO}_2}^s (P - p_{\text{CO}_2}^{\text{sat}})}{RT} \right] \quad (\text{a})$$

Riva et al. (2014) reported that for the determination of solid fugacity in SVE and SLE, various expressions are available in the literature with the assumption that the solid phase consists of a pure freezing component. Soave, (1979) focused on SVE conditions of pure CO₂ by equating pure component fugacities in two phases. This work used SRK EOS to find the liquid and vapor phase fugacities. Since data on sublimation pressure as a function of temperature are known for CO₂, the traditional solid fugacity model can be utilized with the PR, RKS, and NB EOS to explain SVE appropriately. Nasrifar and Moshfeghian. (2020) introduced an SVM model (solid vapor fugacity model) based on studying the CO₂ solid vapor coexistence curve. SVM requires the enthalpy of sublimation, triple point and solid CO₂ molar volume to determine the solid fugacity, and it does not require sublimation pressure. Nasrifar and Moshfeghian. (2022) later developed another model like SVM known as the SLM model (Solid, liquid fugacity model), shown in Eq. b. SLM depended on the enthalpy of melting, triple point and solid CO₂ molar volume. Although SVM provided accurate SVE and SLVE calculations, it was slightly inaccurate for

TABLE 2 Mass transfer model mechanism.

Author	Concept	Phase change method	Description
Wu et al. (2016)	Vapor concentration difference	$\dot{m}_{ai} = \tau_v \cdot \alpha_a \cdot \rho_a \cdot (w_{vi} - w_s(T))$ (1)	τ_v -Time relaxation coefficient
			$\alpha_a \cdot \rho_a$ -Effective density of humid air
Wu et al. (2017)	Gibbs free energy	$\dot{m}_{ai} = \tau_v \cdot (\alpha_a \cdot \rho_a) \cdot w_v \left(\frac{w_v - w_{vs}}{w_{vs}} \right)$ (2)	$(w_v - w_{vs}/w_{vs})$ -Driving force
			w_v - Mass fraction of water vapor
			$\tau_v = 16.705 + 0.0035Re^{1.684}$ (2a)
			$Q_{ia} = h_{ia} (T_i - T_a)$ (2b)
			$h_{ia} = \frac{6\lambda_a \alpha_i \alpha_a N u_{ia}}{d_i^2}$ (2c)
			w_{vs} - Mass fraction of saturated water vapor
			$\alpha_a \cdot \rho_a$ -Effective density of humid air
Sun et al. (2020)	Vapor concentration difference	$\dot{m} = A \cdot h_m \cdot (C_{local,v} - C_{sat,v})$ (3)	A- Contacting area of mass transfer. h_m - Mass transfer coefficient
			$C_{local,v} - C_{sat,v}$ -Mass concentrations of local place and saturated water vapor
			$h_m = \frac{Sh \cdot D_v}{l}$ (3a)
			Sh-Sherwood number
Byun et al. (2020)	Gibbs free energy theory	$\dot{m} = \tau_s \cdot Ste \cdot (\alpha_a \cdot \rho_a) (w_a - w_s)$ (4)	Ste- Stefan number
			τ_s - Time relaxation factor h_{sub} - Latent heat of sublimation
			$Ste = \frac{C_p (T_f - T_w)}{h_{sub}}$ (4a)
Qi et al. (2021)	Temperature difference	$\dot{m} = \lambda_v \alpha_g \rho_g \omega_v df$ (5)	λ_v - Time relaxation coefficient
			Re-Reynolds number. df - Phase change driving force
Haddad et al. (2014)	Concentration difference	$\dot{m}_{CO2,surf} = h_m \cdot A \cdot \rho_{flue\ gas} \cdot (x_{CO2/2} - x_{surf})$ (6)	h_m - Mass transfer coefficient
			$h_m = h_c / \left(\rho_{flue\ gas} \cdot C_{p\ flue\ gas} \cdot Le^{\frac{2}{3}} \right)$ (6a)
			$h_c = \frac{0.21 \cdot Re_{Dh}^{0.8} \cdot P_r^{0.6} \cdot k_{flue\ gas}}{D_h}$ (6b)
			$Le = \frac{k_{flue\ gas}}{\rho_{flue\ gas} \cdot C_{p\ flue\ gas} \cdot D_{flue\ gas}}$ (6c)
			$\dot{Q}_{fr} = k_{eff,fr} \cdot A \cdot \left(\frac{dT}{dy} \right)_{surf}$ (6d)
$k_{eff,fr} = \frac{0.27 \cdot \rho_{fr}}{2386.037 - \rho_{fr}}$ (6e)			

the SLE. By coupling with an EOS, SLM produced satisfactory predictions of the solid fugacity in SVE, SLE and SLVE calculations.

$$f_{\text{CO}_2}^s(T, P) = f_{\text{CO}_2}^L(T, P) \exp \left[\frac{\Delta H_{\text{tp}}}{RT_{\text{tp}}} \left(1 - \frac{T_{\text{tp}}}{T} \right) - \alpha_{\text{CO}_2} \frac{\Delta H_{\text{tp}}}{R^2 T^2} (P - P_{\text{tp}}) \right] \quad (\text{b})$$

3.1.2 Mass transfer models

The frosting process is unsteady with simultaneous heat and mass transfer. Table 2 shows the different frost model mechanisms by the various researchers. Wu et al. (2016) developed a phase change mass transfer model based on vapor concentration difference with local cooling to predict frost layer growth and densification. The concentration difference and effective density of humid air, along with the time relaxation coefficient (τ_v), give the mass transfer rate as shown in Eq. 1. The driving force for the mass transfer rate is the difference between the partial pressure of water vapor in the humid air and the saturation pressure of water vapor corresponding to the surface temperature of the frost. The value of τ_v varies from case to case. In this work, Wu et al. have considered the value of τ_v to be 10. The frost formation on a flat aluminium plate cooled to 10°C was modelled using Euler multiphase flow model. The model was validated, and it was observed that the variation between the modelled and experimental weights of frost was within the range of -3.2%–3.9%.

Wu et al. (2017) developed a frost model based on Gibbs free energy difference to predict the vapor frost formation and growth. The mass transfer rate from the vapor phase to the ice phase \dot{m}_{ai} (Eq. 2), is dependent on the phase change driving force, water vapor effective density and time relaxation coefficient (τ_v) (Eq. 2a). The driving force is equal to the difference of the Gibbs free energy during phase transition. The heat transfer between the two phases (Eq. 2b) is related to the difference in temperature of the two phases and the heat transfer coefficient h_{ia} (Eq. 2c) (Wu et al., 2017). h_{ia} consists of the Nusselt number, which can be determined using the Ranz-marshall correlation (Ranz and Che, 1952). Compared to experimental results, the model predicted frost weight and thickness on a wavy plate with a deviation of -25% ~ + 20% and -20% ~ + 30%, respectively.

Sun et al. (2020) developed a frost model based on vapor concentration differences under cryogenic conditions. The mass transfer rate for vapor phase change (Eq. 3) consists of the driving force, which is the concentration difference between the local place and saturated water vapor. The equation includes the mass transfer coefficient (h_m) term, and the contacting area. h_m can be calculated using the Sherwood number (Eq. 3a). Two correlations of the mass transfer coefficient, the Ranz–Marshall and Jaluria correlations (Ranz and Che, 1952; Whitelaw, 1981), were used for finding the effects of frost formation under free convection on a vertical plate. For forced convection, the Frossling and Tokura correlations can be employed (Patil, 1988; Tokura et al., 1988). The study shows good agreement between the simulation and experimental results and observed the Ranz–Marshall correlation as best suitable for predicting frost formation under cryogenic conditions.

Byun et al. (2020) predicted frost formation under cryogenic conditions using Gibbs-free energy. The mass transfer rate

(Eq. 4), in which the driving force is the Gibbs free energy reduction which is equal to the degree of supersaturation ($\omega_a - \omega_s$). In this model, a term called Stefan number (Eq. 4a) was defined for representing the temperatures of both cryogenic surfaces and air. The value of τ_s was set to 23.6. The frost density and thickness values obtained through simulation on a flat plate show a better comparison with experimental values. It was also noted that the frost with low density was deposited near the frosted surface, and frost with high density formed near the cryogenic surface.

Qi et al. (2021) developed a frost model based on temperature differences under cryogenic conditions. The model was used to explain the trace water vapor frost on a flat plate in a cooled N₂ gas flow. The mass transfer rate for the phase change (Eq. 5) uses Lee's evaporation and condensation mass transfer model (Lee, 2002), which served as the foundation for the introduction of driving force (df), which is due to the difference in control volume temperature and saturation temperature. The model consists of the time relaxation coefficient λ_v (Eq. 5a) and which depends on the Reynolds number and temperature of the cold surface. They validated the simulation with experimental values from the literature and observed the variation to be less than 7%.

Haddad et al. (2014) modelled CO₂ frost development and growth based on concentration differences under cryogenic conditions. Simulations are done to separate CO₂ from the FG mixture and biogas. A correlation of the heat transfer coefficient (Eq. 6b) proposed by Dietenberger et al. (1979) is used for finding the mass transfer coefficient (Eq. 6a) and the mass flow rate of frosted CO₂. The mass transfer rate (Eq. 6) occurs because of the concentration difference between the frosted surface and the FG. The mass transfer coefficient consists of the Lewis number (Eq. 6c) and which links the heat and mass transfer coefficients. The effective thermal conductivity can be determined using Eq. 6e (Shchelkunov et al., 1986). Dietenberger et al. correlation (Eq. 6b) determines the heat transfer coefficient for flow over a horizontal plate. It was observed that Dietenberger et al. correlation provided a better estimation of CO₂ frost growth.

4 Conclusion

CCUS technology is undoubtedly a highly competitive and feasible option, considering global climate change and the increasingly urgent need for the conservation of energy and emission reduction. Compared to other CCUS technologies, CCC is beneficial for increasing the storage capacity and obtaining CO₂ with high purity.

Of the different technologies experimentally tested, the ECL method proposed by Baxter et al. had the least ECC (0.74 GJ/t CO₂) with a capture efficiency of 90%. Based on thermodynamic modeling, the NLCCT technique has reported much lower ECC but requires experimental validation. For a Co of 6.7%, the NLCCT provided a capture efficiency of 99% with an ECC of 0.63 GJ/t CO₂. However, the requirement of maintaining the system at high pressures in ECL and the need for continuous removal of deposited frost in NLCCT, calls for further advancements in CCC techniques.

Based on the review, it is concluded that the desublimation-based frost formation has its own frost mechanism and characteristics which is different from the condensation-based frost formation. For the prediction of the solid phase in the solid-liquid, solid-vapor and solid-

liquid-vapor equilibrium, the SLM model proposed by Nasrifar was found to be accurate. Considering different frost models, Wu et al. frost model based on vapor concentration difference shows less error percentage (−3.2%–3.9%) than other models. Regarding the mass transfer correlations, the Ranz-marshall mass transfer correlation was found to predict frost under cryogenic conditions accurately. In view of the heat transfer correlations, Diitenberger et al. correlation provided a better estimation of the CO₂ frost growth. The correlation was validated for the CO₂ frost growth on a flat plate. Similarly, these heat and mass transfer models need to be verified for different geometries and conditions. Also, the detailed experimental analysis would lead to a better understanding of the frost formation mechanism, which would eventually lead to the development of accurate models.

Author contributions

AA: data curation and writing-original draft preparation, editing. AS: supervision, English language modification, writing-reviewing and editing. All authors contributed to the article and approved the submitted version.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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