Development of a Thermodynamic Carbon Pump Cycle: A Case Study on Temperature Swing Adsorption Carbon Capture (Postprint)

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Abstract

A critical bottleneck constraining the practical implementation of carbon capture technology is the excessively high energy consumption of capture, while thermodynamics serves as a powerful tool for energy system performance analysis. Grounded in the concept of applying thermodynamic research methods to performance analysis of carbon capture technology, this paper takes temperature swing adsorption carbon capture as an example, follows the sequence of "material properties-process-cold and heat sources-cycle" to establish the Thermodynamic Carbon Pump Cycle (TCPC), and subsequently examines the effects of cycle parameters on total energy consumption and second-law efficiency. The results show that cycle energy consumption is primarily influenced by cycle temperature, adsorbent, and adsorption phase, among other factors; the sensible heat of the adsorption phase accounts for approximately 2% of the total cycle energy consumption; the second-law efficiency ranges from 13.91% to 21.21%, indicating significant energy-saving potential. As a "quantitative ruler" based on thermodynamic concepts, TCPC can conduct performance analysis of carbon capture technology, thereby summarizing the main factors affecting total cycle energy consumption, evaluating technology maturity through second-law efficiency, and effectively unlocking the energy-saving potential of carbon capture technology.

Full Text

Construction of Thermodynamic Carbon Pump Cycle: A Case Study on Temperature Swing Adsorption for CO Capture

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Abstract: The primary bottleneck limiting the practical implementation of carbon capture technologies is their excessive energy consumption, while thermodynamics serves as a powerful tool for energy system performance analysis. Based on the concept of applying thermodynamic research methods to analyze carbon capture system performance, this paper uses temperature swing adsorption for CO capture as a case study to construct a thermodynamic carbon pump cycle (TCPC) following the sequence of "property-process-heat source-cycle," and subsequently examines how cycle parameters affect total energy consumption and second-law efficiency. The results demonstrate that cycle energy consumption is mainly influenced by cycle temperature, adsorbent properties, and the adsorbed phase, with sensible heat of the adsorbed phase accounting for approximately 2% of total cycle energy consumption. The second-law efficiency ranges from 13.91% to 21.21%, indicating significant energy-saving potential. As a "quantitative ruler" based on thermodynamic principles, TCPC enables performance analysis of carbon capture technologies, facilitates identification of the main factors affecting total cycle energy consumption, and allows assessment of technology maturity through second-law efficiency, thereby effectively uncovering energy-saving potential in carbon capture technologies.

Keywords: thermodynamic carbon pump; carbon capture; carbon pump cycle; temperature swing adsorption; second-law efficiency

Climate change has gradually become a focal point of global attention, with the greenhouse effect of carbon dioxide representing its primary negative contribution. Among various climate change mitigation technologies, carbon capture and storage (CCS) is considered an effective and proactive technical measure [1]. Currently, the main bottleneck for carbon capture technologies lies in their high energy consumption. For instance, the reboiler heat duty for absorption-based capture technologies is approximately 3–4 GJ per ton of CO captured [2]. On the other hand, carbon capture technologies are diverse, including absorption/adsorption methods, membrane separation, and biological approaches.

At present, targeted tools or models for energy efficiency analysis of carbon capture remain in the exploratory stage. The academic community primarily employs four types of models: (1) Gas separation models—House et al. from MIT [3] used such models to investigate empirical relationships between concentration ratio and separation second-law efficiency, while Ruthven from the University of Maine [4] proposed a value function model to evaluate separation process costs. Gas separation models are lumped models with good universality but excessive simplification. (2) Process analysis models—for example, Sanpasertparnich et al. [5] used ASPEN software to simulate absorption-based carbon capture for an 800 MW coal-fired power plant, while Moullec [6] analyzed thermodynamic limitations of carbon capture technologies on power plant efficiency from post-combustion, pre-combustion, and oxy-fuel combustion perspectives. These models are highly case-specific but lack general applicability, essentially following a "case-by-case" approach without capturing universal principles. (3) Life cycle assessment (LCA) or emergy analysis models, which originate from environmental emission analysis of products over large time scales but inadequately capture details of energy and mass conversion characteristics. (4) Carbon pump models—Zhao et al. [7] proposed the concept of thermodynamic carbon pump from a thermodynamic perspective, introduced its fundamental theory, conducted second-law performance comparisons across five carbon capture technologies, and summarized the maturity of representative technologies.

Compared to the high energy consumption of absorption-based carbon capture, adsorption-based technologies offer distinct advantages, including lower regeneration heat duty, lower required heat quality, higher capture capacity per unit, and fewer system components. Based on how adsorption capacity changes during desorption, adsorption carbon capture technologies are categorized into temperature swing adsorption (TSA) and pressure swing adsorption (PSA). In recent years, numerous scholars have investigated CO adsorption materials [8-10] and adsorption processes [11,12]. Ben-Mansour et al. [13] reviewed physical adsorption of CO from the perspectives of adsorption materials, numerical simulation, and experiments, noting that thermodynamic analysis (both first-law and second-law) should be emphasized for adsorption-based carbon capture.

In summary, energy efficiency analysis models for carbon capture need to be refined to the cycle level for in-depth analysis, and researchers in adsorption carbon capture have recognized the importance of thermodynamic studies. The convergence of these two aspects compels us to consider a critical question: How can we construct a thermodynamic carbon pump cycle using adsorption carbon capture as an example? To address this, this paper focuses on a simple four-step indirect heat exchange TSA technology, conducts detailed cycle construction within adsorption equilibrium diagrams, and analyzes the effects of four factors—desorption temperature, desorption partial pressure, unused bed percentage, and adsorption partial pressure—on total energy consumption and second-law separation efficiency based on thermodynamic carbon pump theory.

1 Cycle Construction

1.1 Methodology

Thermodynamics is an important branch of physics that primarily investigates heat-work conversion relationships, with its analytical objects recently expanding to generalized energy systems. Thermodynamics possesses a mature, self-consistent theoretical framework (the laws of thermodynamics) and precise, powerful analytical tools (exergy, entropy, etc.). When examining performance issues in carbon capture processes, analysis still follows the classic research framework shown in [Figure 1: see original paper]. Using adsorption carbon capture as an example, the foundation of thermodynamic carbon pump (TCP) cycle construction lies in material properties, primarily including adsorbent materials and gas adsorption phases. Subsequently, based on adsorption isotherm equilibrium data and specific heat properties, reasonable thermodynamic processes are designed and "positioned" with heat and cold sources. Multiple processes are then connected to form a complete cycle. Typical thermodynamic processes include heat absorption/release and adsorption/desorption, while typical heat sources include medium-low temperature solar thermal and steam extraction from power plants. Ultimately, cycle construction serves the core objective—energy efficiency analysis—which aligns with the original motivation behind classic thermodynamic concepts such as heat engines and heat pumps, namely to examine process or cycle efficiency.

1.2 Material Properties

The adsorbent used in this TSA carbon capture technology is an amine-functionalized chemically modified adsorption material, with its adsorption isotherm data obtained from Wurzbacher et al.' s experimental work [14]. The Toth model is employed to fit the experimental CO adsorption capacity data, with the detailed formula as follows:

Where P_{CO } is the partial pressure of CO during adsorption, T is the adsorption temperature, H is the heat of adsorption, C is the specific heat, and B , t , D, n , and A are fitting parameters. Other fitting parameters of the Toth model and adsorbent physical parameters are summarized in .

1.3 Process

The research object of this paper is a four-step indirect heat exchange TSA carbon capture technology, with the four steps illustrated in [Figure 2: see original paper]. The four steps are adsorption (AD), preheating (PH), desorption (DE), and precooling (PC). The flue gas to be treated from power plants consists primarily of nitrogen and CO . During adsorption, low-temperature flue gas flows into the adsorption chamber, where CO is selectively adsorbed by the adsorbent material, allowing nitrogen-enriched gas to exit from the other side. During preheating, the adsorption bed is heated by a high-temperature heat source. In the desorption step, CO is released from the adsorption bed under high-temperature conditions. During precooling, the adsorption bed is cooled to ambient temperature by a low-temperature cold source. Through the series connection of these adsorption and regeneration processes, CO separation from flue gas is ultimately achieved.

1.4 Heat and Cold Sources

Adsorption isotherm equilibrium lines determine the upper and lower limits of adsorption and desorption capacity for carbon capture, but are also constrained by external heat and cold source temperatures. Preheating and precooling achieve temperature changes in the adsorption bed through indirect heat exchange, as shown in [Figure 3: see original paper]. Since desorption energy consumption is relatively high, medium-low temperature solar thermal sources can replace steam extraction from coal-fired power plant turbines, limiting the temperature to 343 K. The adsorption process temperature is ambient, with the adsorption isotherm equilibrium line at 296 K as shown in [Figure 3: see original paper].

1.5 Cycle

Corresponding one-to-one with the state points shown in [Figure 3: see original paper], [Figure 4: see original paper] presents the thermodynamic carbon pump cycle diagram in thermodynamic parameter coordinates. For clarity and following conventional expressions in adsorption refrigeration and energy storage, logarithmic coordinates are used. The cycle construction must first satisfy the following assumptions: (1) The temperature within the adsorbent material is considered uniform during each cycle step; (2) Nitrogen adsorption is negligible, and the specific heat of the adsorbed CO phase is calculated using gaseous CO properties; (3) The concentrations of adsorbent material and CO in the adsorption chamber are assumed to reach equilibrium in each cycle.

[Figure 2: see original paper] provides the specific indirect heat exchange TSA carbon capture processes. [Figure 3: see original paper] shows the four steps of the TSA technology within the adsorption isotherm equilibrium boundaries. [Figure 4: see original paper] displays the cycle in logarithmic thermodynamic parameter coordinates. The detailed descriptions of the four specific process steps are as follows:

(1) Adsorption Process 1-2: As shown in [Figure 4: see original paper], the cycle starts at point 1. Power plant flue gas at total pressure 0.1 MPa, CO volume fraction of 10%, and temperature 296 K enters the adsorption chamber. The adsorbent material selectively adsorbs CO , allowing nitrogen-enriched gas to exit the other side. This process is assumed to complete instantaneously. Additionally, adsorption occurs at constant temperature, with the released heat of adsorption removed by cooling water. As shown in [Figure 3: see original paper], the cycle operates between 296 K and 343 K. The theoretical working capacity for CO capture is the difference in adsorption capacity between points 5 and 1. However, due to bed utilization differences, the actual cycle may not reach the equilibrium saturation adsorption capacity under isothermal conditions (the saturation point is at point 5, while the actual adsorption process endpoint is assumed at point 2). The working capacity (*WC*) and unused bed percentage (_{unused}) are defined as follows:

(2) Preheating Process 2-3: Steam is introduced into the heat exchange tubes to heat the adsorption bed while nitrogen purge gas flows through the bed. The CO partial pressure in the bed gradually increases from *P*_{CO ,ad} (point 2) to *P*_{CO ,de} (point 3). During preheating, the adsorption capacity at this partial pressure decreases as temperature rises. When the temperature reaches T , the actual adsorption capacity in the bed equals the equilibrium adsorption capacity at that temperature, meaning the actual adsorption capacity remains unchanged during this stage.

(3) Desorption Process 3-4: As the adsorption bed temperature continues to increase (above T), the actual adsorption capacity gradually exceeds the equilibrium adsorption capacity at the corresponding temperature, causing adsorbed CO to be released from the bed and achieving CO separation. Limited by the high-temperature heat source, the desorption process reaches the maximum temperature T . This desorption process continues with purge gas, maintaining a constant desorption partial pressure.

(4) Precooling Process 4-1: When the adsorption bed temperature reaches the high temperature T (343 K), the product gas line is closed. Cooling water enters the heat exchange tubes, and the adsorption bed temperature begins to decrease, gradually dropping to ambient temperature T (296 K). No CO adsorption or desorption occurs during this process, meaning the actual adsorption capacity in the bed remains constant. The cycle returns from point 4 to point 1.

2 Thermodynamic Carbon Pump Evaluation Parameters

Generally, CO separation performance is evaluated using CO recovery rate (R_{CO }) and product gas purity. Since nitrogen adsorption is not considered in this paper, product gas purity is not used as an evaluation metric. The flue gas flow rate in the adsorption process is designed based on the ideal working capacity (WC_{id}), so the actual recovery rate depends only on the unused bed percentage, defined as follows:

Zhao et al. [7] detailed the fundamental theory of thermodynamic carbon pump, where the minimum separation work (W_{min}) represents the power consumption for reversible separation of an ideal gas mixture under isothermal and isobaric conditions without chemical reactions. Its physical significance is the difficulty of achieving the specified gas mixture separation. The minimum separation work is independent of the separation path and depends only on the initial and final states of the separation process. In carbon capture, the minimum separation work depends solely on three process parameters: separation temperature, initial CO concentration, and recovery rate. Furthermore, the second-law separation efficiency (_{2nd}) is the ratio between the minimum separation work of the ideal process and the net exergy input of the actual process. Therefore, the formulas for minimum separation work and second-law separation efficiency are as follows:

In evaluating the four-step indirect TSA carbon capture process in this paper, the primary energy consumption is the total heat duty of the separation process. Therefore, power consumption from gas blowers and pumps is neglected. Additionally, cooling water temperature is assumed equal to ambient temperature, and temperature differences between heat/cold sources and the adsorption bed are ignored. Consequently, the total energy consumption (Q_{H}) of the four-step indirect heat exchange TSA process includes sensible heat of the adsorbed phase, sensible heat of the adsorbent, and latent heat of adsorption, calculated as follows:

3 Results and Discussion

This paper employs thermodynamic carbon pump theory to analyze the effects of four factors—desorption temperature, desorption partial pressure, unused bed percentage, and adsorption partial pressure—on the separation energy consumption and second-law separation efficiency of indirect heat exchange TSA technology.

3.1 Desorption Temperature

Desorption temperature significantly affects the separation performance of TSA carbon capture technology. Generally, as desorption temperature increases, CO adsorption capacity decreases. As described in the cycle shown in [Figure 4: see original paper], this section analyzes four TCP cycles with different desorption temperatures: 343 K, 348 K, 353 K, and 358 K. All other state parameters remain identical: adsorption temperature of 296 K, adsorption CO partial pressure of 10 kPa, desorption CO partial pressure of 40 kPa, and unused bed percentage of 5%.

[Figure 5: see original paper] presents the variation trends of total energy consumption and second-law separation efficiency for TCPC under the four desorption temperature conditions. As desorption temperature increases from 343 K to 358 K, both ideal and actual working capacities increase, resulting in a corresponding increase in CO recovery rate from 83.48% to 87.32%. According to the minimum separation work calculation formula, with separation temperature and initial concentration held constant, the minimum separation work values are 162.39 kJ/kg, 163.58 kJ/kg, 164.58 kJ/kg, and 165.44 kJ/kg, respectively.

Since the average CO heat of adsorption is used as the latent heat for the cycle, the latent heat values are equal across different desorption temperatures at 1363.63 kJ/kg. Because the increase in actual working capacity outweighs the heat gain from temperature differences, both adsorbent sensible heat and adsorbed phase sensible heat show decreasing trends. Specifically, adsorbent sensible heat decreases from 4858.40 kJ/kg to 4700.94 kJ/kg (a 3.35% reduction), while adsorbed phase sensible heat decreases from 121.28 kJ/kg to 103.45 kJ/kg (a 17.34% reduction). Consequently, total cycle energy consumption shows a decreasing trend, dropping from 6343.31 kJ/kg to 6168.03 kJ/kg. The sensible heat of the adsorbed phase calculated based on gas-phase properties accounts for 1.68%–1.91% of total energy consumption. Although total energy consumption decreases with increasing desorption temperature, the Carnot factor based on exergy calculations increases. Therefore, the second-law separation efficiency calculated by the formula shows a decreasing trend, specifically from 18.68% to 15.49%.

3.2 Desorption Partial Pressure

Generally, the primary factors affecting CO partial pressure during desorption are purge gas (nitrogen) flow rate and temperature. As described in the cycle shown in [Figure 4: see original paper], this section analyzes four TCP cycles with different desorption partial pressures: 35 kPa, 40 kPa, 45 kPa, and 50 kPa. All other state parameters remain identical: adsorption temperature of 296 K, adsorption CO partial pressure of 10 kPa, desorption temperature of 343 K, and unused bed percentage of 5%.

[Figure 6: see original paper] presents the variation trends of total energy consumption and second-law separation efficiency under the four desorption partial pressure conditions. As desorption partial pressure increases from 35 kPa to 50 kPa, both ideal and actual working capacities decrease, resulting in a slight reduction in CO recovery rate from 83.68% to 83.15%. With separation temperature and initial concentration held constant, the minimum separation work values decrease from 162.55 kJ/kg to 162.15 kJ/kg.

Since the average CO heat of adsorption is used as the cycle latent heat, the latent heat values are equal at 1363.63 kJ/kg. As desorption partial pressure increases, T also increases correspondingly, causing adsorbed phase sensible heat to increase from 118.66 kJ/kg to 125.41 kJ/kg. The increase in desorption partial pressure reduces actual working capacity, leading to an increasing trend in adsorbent sensible heat from 4785.77 kJ/kg to 4974 kJ/kg. The adsorbed phase sensible heat calculated based on gas-phase properties accounts for 1.89%-1.94% of total energy consumption. Total energy consumption shows an increasing trend with desorption partial pressure, while the exergy of separation cycle energy consumption increases since desorption temperature remains constant. According to the second-law formula, the decreasing minimum separation work causes second-law separation efficiency to decline from 18.93% to 18.81%.

3.3 Unused Bed Percentage

Factors such as flow velocity differences, insufficient adsorption time, and dead volume within the system affect adsorption bed utilization. As described in the cycle shown in [Figure 4: see original paper], this section analyzes four TCP cycles with different unused bed percentages: 5%, 7%, 9%, and 11%. All other state parameters remain identical: adsorption temperature of 296 K, adsorption CO partial pressure of 10 kPa, desorption temperature of 343 K, and desorption CO partial pressure of 40 kPa.

[Figure 7: see original paper] presents the variation trends of total energy consumption and second-law separation efficiency under the four unused bed percentage conditions. As unused bed percentage increases from 5% to 11%, the ideal working capacity remains constant while the actual working capacity decreases, causing continuous decline in CO recovery rate: 83.48%, 76.87%, 70.26%, and 63.65%, respectively. With separation temperature and initial concentration held constant, the corresponding minimum separation work values decrease from 162.39 kJ/kg to 150.58 kJ/kg.

Since the average CO heat of adsorption is used as the cycle latent heat, the latent heat values are equal at 1363.63 kJ/kg. As unused bed percentage increases, T increases correspondingly, but actual working capacity decreases. Therefore, adsorbed phase sensible heat increases from 121.28 kJ/kg to 162.65 kJ/kg. The reduction in actual working capacity combined with constant temperature difference causes adsorbent sensible heat to increase from 4858.40 kJ/kg to 6341.79 kJ/kg. The adsorbed phase sensible heat calculated based on gas-phase properties accounts for 1.91%-2.06% of total energy consumption. Total energy consumption shows an increasing trend with unused bed percentage, while the exergy of separation cycle energy consumption increases since desorption temperature remains constant. According to the second-law formula, the decreasing minimum separation work causes second-law separation efficiency to decline from 18.68% to 13.91%.

3.4 Adsorption Partial Pressure

Initial flue gas pressure and CO concentration determine the adsorption process CO partial pressure. As described in the cycle shown in [Figure 4: see original paper], this section analyzes four TCP cycles with different adsorption partial pressures: 5 kPa, 10 kPa, 15 kPa, and 20 kPa. The initial flue gas pressure is maintained at 0.1 MPa with CO volume fractions of 5%, 10%, 15%, and 20%, respectively, which falls within the typical range for post-combustion carbon capture. All other state parameters remain identical: adsorption temperature of 296 K, unused bed percentage of 5%, desorption temperature of 343 K, and desorption CO partial pressure of 40 kPa.

[Figure 8: see original paper] presents the variation trends of total energy consumption and second-law separation efficiency under the four adsorption pressure conditions. As adsorption partial pressure increases from 5 kPa to 20 kPa, both ideal and actual working capacities increase, resulting in slight growth in CO recovery rate from 82.25% to 84.28%. According to the minimum separation work calculation formula, with separation temperature held constant and both initial concentration and recovery rate increasing, the minimum separation work values are 201.46 kJ/kg, 162.39 kJ/kg, 138.83 kJ/kg, and 121.65 kJ/kg, respectively.

Since the average CO heat of adsorption is used as the cycle latent heat, the latent heat values are equal at 1363.63 kJ/kg. As adsorption partial pressure increases, T decreases while actual working capacity continues to grow. Therefore, adsorbed phase sensible heat decreases from 138.06 kJ/kg to 110.26 kJ/kg. The increasing actual working capacity combined with constant temperature difference causes adsorbent sensible heat to decrease from 5458.14 kJ/kg to 4477.36 kJ/kg. The adsorbed phase sensible heat calculated based on gas-phase properties accounts for 1.85%-1.98% of total energy consumption. Total energy consumption shows a decreasing trend with adsorption partial pressure, while the exergy of separation cycle energy consumption decreases since desorption temperature remains constant. According to the second-law formula, the increasing minimum separation work causes second-law separation efficiency to decline from 21.21% to 14.92%.

[Figure 9: see original paper] presents the energy efficiency analysis of carbon pump cycles for different adsorption technologies. PSA and electric swing adsorption (ESA) technologies primarily consume work (electricity) from external sources, while TSA technology consumes heat. The figure reveals that, except for ESA, existing adsorption carbon capture technologies achieve second-law separation efficiencies between 10% and 25%, though energy consumption amounts differ due to variations in energy quality and adsorption materials. For TSA, the modified adsorbent used in this study is suitable for relatively low desorption temperatures, making it appropriate for integration with medium-low temperature solar thermal or waste heat sources, thus offering energy-saving potential despite its relatively high heat consumption and efficiency similar to other studies. However, ESA technology uses electricity to generate heat for CO separation, essentially representing a special type of TSA technology. Consequently, its total energy consumption is similar to TSA technology, but its efficiency is much lower due to energy quality differences. This figure demonstrates that, unlike research methods in chemical engineering, TCP cycle construction can quickly and accurately outline the efficiency characteristics of a class of carbon capture technologies, enabling both vertical comparisons within a single technology domain and horizontal comparisons across representative technologies as shown in reference [7]. Similar to the significance of previous work on heat engines and heat pumps, this contribution lies in fully leveraging thermodynamics as a "quantitative ruler" for energy systems, effectively guiding engineering practice, reasonably quantifying technology roadmaps, eliminating blind technology development, and safeguarding efficient industrial technology advancement.

Conclusions

This paper proposes a method for constructing cycles within adsorption isotherm equilibrium diagrams for a four-step indirect heat exchange TSA carbon capture technology, and uses thermodynamic carbon pump theory to evaluate the effects of desorption temperature, desorption partial pressure, unused bed percentage, and adsorption partial pressure on total energy consumption and second-law separation efficiency. The following conclusions are drawn:

(1) As a thermodynamic analysis method and tool, TCPC can accurately address energy efficiency issues in carbon capture processes. Compared to traditional gas separation models, it is more specific, and compared to process analysis models, it is more universal, possessing clear characteristics

as a “quantitative ruler” for energy systems.

(2) Based on the classic thermodynamic progressive research approach of “property-process-heat source-cycle,” it can be concluded that calculating the adsorbed phase CO properties using gas-phase properties results in sensible heat of the adsorbed phase accounting for approximately 2% of total cycle energy consumption.

(3) The total energy consumption of adsorption technology primarily depends on cycle construction, adsorbent properties, and adsorbed phase properties. Energy consumption is dominated by adsorbent sensible heat and desorption latent heat, with adsorbent sensible heat exceeding desorption latent heat.

(4) Based on TCP cycle analysis, the indirect heat exchange TSA technology using amine-modified adsorbent for CO separation achieves second-law separation efficiencies between 13.91% and 21.21%. The second-law separation efficiency decreases correspondingly when desorption temperature, desorption partial pressure, unused bed percentage, and adsorption partial pressure increase.

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