Research Article

Optimization of the Natural Gas Purification Process Based on Exergy Analysis

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This study is aimed at carrying out investigations on a domestic gas field, located in Yanchang, China, with a view to optimize the natural gas purification process. The main objectives of this work are (i) to reduce the natural gas purification system’s energy consumption and (ii) improve the existing purification levels. Process simulations were carried out using Aspen Plus™ software, and a comprehensive technical and economic analysis was carried out. The single-factor sensitivity analysis method was used to determine the parameters of absorption, such as the reflux ratio and number of stages. The heat transfer process was analyzed using the energy-saving method of the energy system, and a modified process was recommended. The optimization results show that the recommended system has better purification performance, the comprehensive energy consumption is effectively reduced, and the energy efficiency is improved by 9%.

1. Introduction

To promote a healthy, sustainable, and stable development of our national economy, the energy requirements of high-energy-consuming systems should be reduced. Therefore, improving the energy utilization rate has become one of the main research directions. In the field of natural gas purification, high-energy consumption and complex processes are widely prevalent [1], which makes it a potential candidate for carrying out an energy-saving analysis and reducing the energy consumption. Most of the studies carried out in the past only analyzed the process flow from the perspective of a single aspect, which usually resulted in a limited degree of optimization. Thus, the optimization results were inadequate to meet the needs of the industry, and the benefits of the transformation were not obvious. Based on a summary of the optimization schemes employed in the previous works, a multidimensional optimization and a comprehensive and systematic analysis of natural gas purification systems have been conducted in this study from the perspectives of process and technology. This study was conducted at the Yanchang gas field in China. The first step involved the selection of a deacidification reagent. The current solution systems include methyl diethanolamine (MDEA) solution, MDEA complex solution, activated MDEA solution, MDEA-sulfolane solution, and MDEA mixed solution. The second step involved the improvement of the deacidification process, for which there were two options, the poor/rich liquid circulation system and the semipoor liquid circulation system. The third step involved the modification of the dehydration process through low-temperature separation, solvent absorption, solid absorption, supersonic dehydration, and membrane separation. The final step involved the optimization of the heat exchanger network from the perspective of exergy analysis, which not only improves energy efficiency but also saves energy.

2. Optimization of the Natural Gas Purification Process Based on Exergy Analysis

2.1. Deacidification Process Optimization
2.1.1. Reagent Selection. The common acid gas components in the natural gas industry are CO₂ and H₂S. For meeting a variety of requirements, such as environmental protection from exhaust gas emissions, protection of equipment and pipelines from corrosion, and downstream processes, H₂S gas must be removed from the systems. It should be noted that the requirements for the allowable H₂S content are very stringent. However, CO₂ is mainly responsible for greenhouse gases, which have serious consequences for the environment. CO₂ recovery and emission reduction are the main methods to mitigate the greenhouse effect [2]. Macroscopically speaking, acid gas deacidification can be divided into two methods, chemical absorption and physical adsorption. The sulfur part of the Sulige gas field is located in the Lower Paleozoic horizon, mainly Taiyuan Formation and Mawu Formation, while the gas well horizon of the Yanchang oilfield includes Benxi Formation, Shanxi Formation, and Shihezi Formation of Upper Paleozoic, so it does not contain sulfur and is absent in the natural gas processing components as well but has relatively low gas source pressure and high-water content. Therefore, it has higher treatment requirements in the downstream treatment. Hence, the only other main component of acid gas is CO₂. The acid gas of the original treatment plant was removed by the chemical absorption method. The reagent used was MDEA solution, i.e., aqueous N-methyl diethanolamine. MDEA is a tertiary amine, and its deacidification mechanism is shown in formulas (1)–(3) [3–5].

\[
\text{CO}_2 + \text{R}_2\text{NCH}_3 \rightarrow \text{nonreactive} \quad (1)
\]

\[
\text{CO}_2 + \text{H}_2\text{O} \rightarrow \text{H}^+ + \text{HCO}_3^- \quad (2)
\]

\[
\text{H}^+ + \text{R}_2\text{NCH}_3 \rightarrow \text{R}_2\text{NCH}_3\text{H}^+ \quad (3)
\]

MDEA is a reagent for selective absorption of sulfur in natural gas purification pretreatment in use since the early 1980s. It has many advantages, such as good degradation performance, great chemical stability, long shelf life, low circulation of amine liquid, and relatively low price [6]. However, MDEA foams more easily than the other amine liquids and, hence, can often lead to false liquid levels in the absorption and regeneration towers. Furthermore, it is not conducive for personnel to operate and manage the process and can, therefore, even cause serious safety issues. To improve the stability of the MDEA system and comply with the principle of selecting reagents according to the CO₂ concentration in engineering [7], two suitable optimization schemes are proposed for this system:

1. The compound amine solution is used for deacidification. Compound amine solution has many advantages, such as ability to improve purification, high absorption load capability, capacity for fast absorption of the acid gas, lower levels of corrosion of equipment and pipelines, and low energy consumption of regeneration and low evaporation loss. It makes up for the shortcomings of the single amine solution [8, 9]. The compounding scheme used in this study is based on the previous literature [10], with a specific combination of 2 mol/L MDEA with sulfolane in a ratio of 10:3 to create a complex amine liquid.

2. Because there is no active H atom in the MDEA molecule, its reaction must be based on the reaction of CO₂ with H₂O (control step) [11]. Thus, its absorption of CO₂ can be improved by activating the MDEA method [12]. By adding primary amine, secondary amine, or another activator to the MDEA solution, this method can achieve high acid gas load and low energy consumption. Taking primary amine as an example, the reaction mechanism is shown in (4)–(6) [13]:

\[
\text{CO}_2 + 2\text{RNH} + \text{H}_2\text{O} \rightarrow \text{RNHCOO}^- + \text{RNH}_2^+ \text{(medium speed reaction)} \quad (4)
\]

\[
\text{CO}_2 + \text{RNH} + \text{H}_2\text{O} \rightarrow \text{RNH}_2^+ + \text{HCO}_3^- \quad (long \ response) \quad (5)
\]

\[
\text{RNHCOOH} + \text{RCH}_3\text{NH} + \text{H}_2\text{O} \rightarrow \text{RNH} + \text{RCH}_3\text{NH}^+ \cdot \text{HCO}_3^- \quad (6)
\]

The abovementioned reactions show that the activator forms carboxylic acid after absorbing CO₂ and then immediately transfers it to the liquid phase to form bicarbonate. In this process, the activator acts as a catalyst without consumption [14]. Therefore, an activator is very commonly used in natural gas decarbonization treatment. Using the experience of the past studies [15, 16], in this study, PZ (piperazine) with a superior activation performance is used as the activator. This activator has strong adaptability and can meet the requirements of CO₂ absorption at different concentrations. This is because in the later stage of the gas field exploitation, as the availability of the natural gas reserves decreases, it is accompanied by an increase in the CO₂ content. Therefore, the selection of an activator with a wide range of sensitivity has a positive significance for gas treatment in the later stage of the gas field. The mass fraction of PZ in the final selected activated MDEA solution is 3%–5%.

2.1.2. Process Improvement. By studying the influence of several process parameters on the CO₂ content in the purified gas and the operational cost of the unit, it is concluded that the absorption temperature of the amine solution, circulation volume, and regeneration tower temperature all have a significant influence on the operational cost of the unit [17]. Of these three, the most influential factors are the circulating quantity of the amine solution and the regeneration temperature. A small change in these two parameters would cause the operational cost of the unit to increase significantly. Thus, the superiority of the semilean solution scheme [18] (wherein half of the total circulating solution is extracted from the middle of the regeneration tower, after passing through the semilean liquid/rich liquid heat transfer, and is reintroduced to the middle of the absorption tower) can be evident.
2.2. Dehydration Process Optimization. In addition to solid and acid gases, impurities in raw natural gas also contain water. The existence of water increases the risk of hydrate formation, causes ice blockage of pipelines and equipment, affects the calorific value of the natural gas, reduces the quality of natural gas, and wastes energy in the downstream treatment [19]. Therefore, dehydration of natural gas is very essential. There are two methods for water treatment, chemical absorption and physical absorption [20] (membrane separation [21, 22], silica gel and molecular sieve method [23]). To improve the recovery of natural gas in the later stage of gas field exploitation, pressurized foam drainage or fracturing technology is generally adopted, which will inevitably lead other pollutants such as reagents, oil pollution, and gas field water into the purification process, thus affecting the dehydration effect. Secondly, the increase of turbocharging will cause airflow pulsation, which will bring great hidden dangers to the safe production of natural gas purification. Moreover, the consumption of triethylene glycol (TEG) in the TEG dehydration process will account for more than half of the dehydration process and become an important factor affecting the operation cost of the dehydration process [24]. At the same time, considering that most of the natural gas output of the Yanchang gas field will be sent to an LNG processing plant, a three-tower molecular sieve dewatering process is recommended to meet the high standards of the LNG gas source and the relevant requirements of storage and transportation in the health, safety, and environment (HSE) management system [25, 26]. A typical three-tower molecular sieve dehydration process is available in the literature [27]. The shadow part shows that the water content is basically saturated, and the blank area shows that water can also be adsorbed.

2.3. Exergy Analysis. The first law of thermodynamics states that energy is conserved when it is transformed or transferred, and the second law of thermodynamics states the degree and direction of the process. Exergy analysis combines the first and second laws. It is a scientific energy analysis method of energy systems. It plays a unique role in the global energy-saving practice [28, 29]. Exergy refers to the maximum functional power [30] when the system is in a certain state and can be expressed as

\[ E_x = (H - H_0) - T_0(S - S_0), \]  

where \( T_0 \) is the ambient temperature, \(^\circ\)C; \( H \) is the specific enthalpy, kJ; \( H_0 \) is the specific enthalpy under ambient temperature, kJ; \( S \) is the entropy, kJ; and \( S_0 \) is the entropy under ambient temperature, kJ.

In recent decades, the modern energy use theory, which takes the method of exergy analysis as the core, has been widely used in energy management, petrochemical industry, thermal power plants, refrigeration technology, and other fields. It provides a scientific basis for further improving the level of energy use and effectively reducing the energy consumption index. There are mainly three kinds of exergy analysis models [31]. Generally, the black box and gray box models are mostly used for qualitative analysis of the energy consumption system, and then the fuzzy analysis results are used to carry out energy-saving transformation. However, the energy consumption data of each unit of the system is complete, and the white box model analysis can ensure the accuracy of the data, so as to obtain a better transformation effect, so the white box model is used in this paper.

2.4. Heat Exchanger Network. Since the 1960s, heat exchanger network integration technology has attracted significant attention as a subproblem of process design [32]. The transformation of the heat exchanger network effectively avoids the problems of temperature crossover and reverse heat transfer [33]. In 1965, Hwa first proposed structural optimization of heat exchanger networks [34]. In 1970, Ponton and Nishia proposed a trial method [35]. Shen et al. and Linnhoff et al. [36, 37] proposed the pinch analysis method, which is a method to solve the problem of inconsistency of energy flow in heat exchanger networks [38].

3. Comparison of Simulation Results between the Actual and Optimized Processes

3.1. Overview of Actual On-Site Processes. The purpose of process simulation is to realize an optimal design, operation, and modification of the model or system in a relatively short time by changing the relevant parameters in the model. The PR (Peng-Robinson) equation suitable for oil and gas processing units is chosen as the state equation for simulation. The natural gas components are shown in Table 1. The process simulation model in Aspen Plus is depicted in Figure 1.

3.2. Optimization Results

3.2.1. Selection of the Deacidification Reagent. The original process is optimized step by step. Firstly, the deacidification reagent is selected, and the reagent type is changed at flow 134 for simulation. In the first simulation, the original site solution is added to only 3 mol/L MDEA solution. In the second simulation, 2 mol/L MDEA and sulfolane are added to the compound amine solution with a molar ratio of 10:3. In the third simulation, 3% PZ is added in 2 mol/L MDEA solution. A comparison of the results is shown in Table 2.

It can be easily seen that the purification effect of the activated MDEA solution is slightly better than that of the compound amine solution and it achieves the purpose of deep

| Components | Mole percent (%) | Components | Mole percent (%) |
|------------|------------------|------------|------------------|
| He         | 0.0314           | CH₂=CH₂       | 0.0214           |
| H₂         | 0.0152           | CH₃-CH₃       | 0.0015           |
| N₂         | 0.2698           | CH₃=CH₂–CH₂–  | 0.0023           |
| CO₂        | 2.9078           | C(CH₃)₄      | 0.0005           |
| CH₄        | 96.105           | (CH₃)₂CHCH₂CH₃ | 0.0006           |
| CH₃-CH₄    | 0.5676           | Hg           | 1                |
purification, which is more amenable to the follow-up LNG processing process. Hence, this method is recommended for further analyses.

3.2.2. Absorption Tower Parameters. Simple calculation results of the absorption tower are shown in Table 3.

3.2.3. Semilean Liquid Process. To reduce the circulation of the absorption tower and the regeneration temperature of the regeneration tower, the optimized process uses a semilean solution scheme, as shown in Figure 2. To determine the entrance position of the semilean liquid, the number of trays can be increased appropriately to find out the rule. Therefore, sensitivity analysis of the absorption tower is carried out as shown in Figure 3. The Y axis represents the gas phase molar fraction of CO$_2$ and CH$_4$ at different positions in the absorber, and the X axis represents the number of theoretical trays (the first stage at the top of the absorber). By analyzing the distribution curves of key components in the absorber, it can be seen that the absolute values of the slope of CO$_2$ and CH$_4$ curves begin to decrease at 3/4 and 1/3 of the distance from the bottom of the absorber, indicating that

| Methods                  | MDEA solutions | Compounded MDEA solutions | Activated MDEA solutions |
|--------------------------|----------------|---------------------------|--------------------------|
| Postpurification CO$_2$ content (mol%) | 0.0041 (the field observation values) | 0.0023 | 0.0018 |

| Items                              | Value       |
|------------------------------------|-------------|
| Absorption pressure                | 5720 (kPa)  |
| Absorption temperature             | 35 (°C)     |
| Gas-liquid ratio                   | 423 (m$^3$/m$^3$) |
| Number of actual stages            | 15.3522     |
the absorption efficiency decreases here. To maintain high absorption efficiency for a longer duration, it is recommended to introduce semian liquid at 3/4 or 1/3 of the whole column.

3.2.4. Three-Tower Molecular Sieve Dehydration Process. In the traditional two-tower molecular sieve dewatering process, tower A is for the adsorption operation and tower B is for the regeneration of adsorbents. However, in the three-tower and multitower processes, owing to various factors, switching procedures between different towers can be selected. Taking a three-tower process as an example, towers A, B, and C can be used for adsorption, regeneration, and cooling, respectively. Alternatively, towers A, B, and C can also be used for regeneration, adsorption, and cooling, respectively. Irrespective of these choices, the basic principles remain the same. The three-tower molecular sieve dewatering process has strong independence. It can not only save power but also eliminate a set of regenerated gas turbocharging units. A typical three-tower process is shown in Figure 4.

3.2.5. Optimization of the Heat Exchanger Network. Aspen Energy Analyzer is used to analyze the relationship between the operational cost, equipment cost index, and minimum heat transfer temperature difference. Considering the actual heat transfer effect and the goal of minimizing the total investment, the pinch point method is used to analyze the minimum heat transfer temperature difference of this project, which is 13°C, as shown in Figure 5.

Based on the premise of the minimum heat transfer temperature difference, the heat exchanger network is optimized. The specific optimization details are shown in Figure 6.

Specific optimization steps are as follows: the acid gas at the top of the amine liquid regeneration tower carries a large amount of latent heat at low temperatures, which cannot be directly used. Heat pump technology can be used to raise the acid temperature level and then be used as the heating heat source of the bottom reboiler to raise the acid temperature level and then be used. Heat pump technology can be used to extract a large amount of latent heat at low temperatures, which matches the temperature and the heat load of the refrigeration cycle cooler perfectly during pressure swing adsorption operation, thus avoiding the use of process logistic heat transfer but using the circulating water for cooling. Similarly, the heater should adopt steam heating to ensure the stability of the device. Through the optimization of the heat exchanger network, a total of nine heat exchanger flows are realized, and all the parts that need to be optimized are reflected in the simulation process. The process is transformed and constructed according to the optimization results. The improved simulation process is shown in Figure 7.

3.2.6. Exergy Analysis. The main components of the purification plant are the MDEA solution circulation system, triethylene-glycol circulation system, production and fire water supply system, air-nitrogen system, boiler and boiler water supply system, sewage treatment system, and circulating water system. By comparing the energy consumption data of the components, it is found that the dominant power consumption components are the circulating pump of the MDEA circulating system and the air compressor of the air-nitrogen system. The power consumption details of these units are shown in Table 4.

The formula for the enthalpy loss in the heat exchanger is as follows, wherein 1 and 2 are the states before and after the heat exchange.

\[ \Delta E_x = (H - H_0) - T_0(S - S_0) + \omega_c, \]  

where \( T_0 \) is the ambient temperature, °C; \( H \) is the specific enthalpy, kJ; \( H_0 \) is the specific enthalpy under ambient temperature, kJ; \( S \) is the entropy, kJ; \( S_0 \) is the entropy under ambient temperature, kJ; and \( \omega_c \) is the compressor power consumption, kW.

The formulae for calculating the rate loss in the heat exchangers are as follows, 1 and 2 of which are pre- and post-heat transfer states.

\[ \Delta E_x = \sum E_{x,in} - \sum E_{x, out}, \]

where \( E_{x,in} \) is the exergy entering the heat exchanger, kJ; and \( E_{x,out} \) is the exergy leaving the heat exchanger, kJ.
According to the process flow of the device and considering the main energy-consuming process equipment, the model for the analysis is established as shown in Figure 8. The results of each logistic point in the plant under two operating conditions are shown in Table 5. Because the purpose of this study is to provide a basis for decision-making for the operation of the plant, additional fuel and other factors are needed in the analysis and calculation. The equations of equilibrium are shown as follows.

\[ E_1 c_1 + E_4 c_4 = E_2 c_2 + E_3 c_3, \]
\[ E_3 c_3 + E_5 c_5 + E_{11} c_{11} + E_{12} c_{12} = E_4 c_4 + E_6 c_6 + E_{14} c_{14}, \]
\[ E_6 c_6 + E_8 c_8 + E_{10} c_{10} = E_5 c_5 + E_7 c_7 + E_9 c_9, \]
\[ E_7 c_7 + E_8 c_8 + E_{14} c_{14} = E_9 c_9 + E_{10} c_{10} + E_{11} c_{11} + E_{15} c_{15} + E_{18} c_{18}, \]
\[ E_{15} c_{15} = E_{17} c_{17} + E_{18} c_{18}, \]

where \( E_x \) is the exergy value of \( x \), kJ; and \( c_x \) is the unit cost of \( x \), RMB yuan/kJ.

There are a total of 18 unknowns and only 5 economic equilibrium equations. To solve the equations, auxiliary equations are established as follows:

1. The unit costs of the inlet and outlet of the compression unit and the dry gas of the outlet are equal, i.e., \( c_6 = c_7 = c_{14} \)
2. The unit costs of the terminal products are equal, i.e., \( c_{17} = c_{18} \)
3. The raw material gas is close to the environmental state, and its unit cost is zero, i.e., \( c_{12} = 0 \)
4. Fuel gas physical exergy is neglected. The gas used in the fuel gas system comes from a part of the produced gas of the well plant, and it is converted into atmospheric gas for combustion after passing through the throttle needle valve and pressure-reducing valve, and the temperature is normal temperature.
Therefore, in order to make the calculation simple and the equation have a unique solution, it is ignored.

(5) The unit cost of dry gas is calculated according to the following formula:

$$c = \frac{e_{13} \times E_{13} + (Q - Q_{13}) \times e_{17} \times e_{17}}{E},$$

where $Q$ is the flow of dry gas, m$^3$; $Q_{13}$ is the flow of node 13, m$^3$; and $e_{17}$ is the specific energy of node 17.

The enthalpy loss of the compression process in the original process flow is calculated to be 32 MJ/h, and the enthalpy loss in the heat exchange process is 46.2 MJ/h. After optimization, the process enthalpy loss is 29.04 MJ/h and the heat exchange enthalpy loss is 42.715 MJ/h. This translates to an increase of efficiency by approximately 9%. Thus, the expected energy-saving goals have been achieved.

4. Conclusion

(1) As per the data from the single-factor analysis and the Aspen Plus simulation, the purification rate of CO$_2$ by MDEA with PZ as an activator reached 99.94%, which is significantly higher than that by the MDEA-sulfolane mixed solution method. While ensuring the purification effect, the semilean ammonia liquid circulation process reduced the ammonia liquid circulation by more than 49% compared with the ordinary lean/rich ammonia liquid circulation process. Furthermore, this also reduced the regeneration load of the ammonia liquid regeneration tower and achieved the goal of energy saving. Using a three-tower molecular sieve in the optimized dehydration process can not only avoid the investment cost caused by the TEG loss but also save 583,500 yuan per year for the plants based on a consumption of 14.03 mg of triglyceride per m$^3$ of natural gas treatment. At the same time, it can meet the temperature requirements of downstream LNG production.

(2) Based on the analysis of the heat exchanger network, the pinch analysis method is used to optimize the heat exchanger network of the optimized purification process. From the thermodynamic point of view, the optimal matching of the heat exchanger logistics and the thermodynamic goal of minimum energy consumption are achieved. In addition, the problem of inconsistency of energy flow in the heat exchanger network is solved. Finally, the efficiency of the system is improved. Thus, all the goals of optimization have been achieved.

Data Availability

The data used to support the findings of this study are available within the article.
Conflicts of Interest

The authors declare that they have no conflicts of interest.

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