

Low-temperature purification of the synthesis gas

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Abstract. In this paper, studies are made of the most economical method of low-temperature purification of synthesis gas, which is obtained during steam gasification of carbonizate. The analytical calculation is based on the cooling and phase transition of sulfur and carbon dioxide compounds and their separation. The paper presents the comparison of process energy costs depending on consumption characteristics and process parameters.

1 Introduction

Synthesis gas obtained in the process of steam oxygen-free gasification [1] is characterized by the content (% by vol.) of the target H₂ components equal to 62, while CO - to 26. In addition, it contains gases such as CO₂ up to 12, H₂S and SO_x up to 1% by vol., which adversely affect the catalytic processes of obtaining various hydrocarbons. For the Fischer-Tropsch process, most catalysts, including iron and cobalt, are extremely sensitive to "poisoning" with sulfur that is reflected in the shortening of their working life and deterioration of the process selectivity. As a result, it is impossible to reach the calculated technical and economic indicators and to ensure a low cost price of the products. In this connection, the most important is the purification of synthesis gas from sulfur compounds (SO_x and H₂S) and CO₂. Despite the relatively low concentration of sulfur compounds in the composition of synthesis gas (up to 1% by vol.) their content should be ensured up to 2 mg/m³ [2].

2 Statement of the problem

The purpose of these studies is the energy analysis for the proposed method of purification of synthesis gas produced in the steam gasification carbonizate. It is proposed to purify the synthesis gas by selective desublimation/condensation of the components by discrete cooling. The implementation of such a method can compete with chemical methods of purification.

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3 Process and schema description

The gas is cooled to the phase transition temperature of its components at atmospheric pressure: CO_2 - below 95K, H_2S - below 213K.

In order to investigate the physical methods of purification of synthesis gas, we used a principal scheme, shown in Fig. 1.

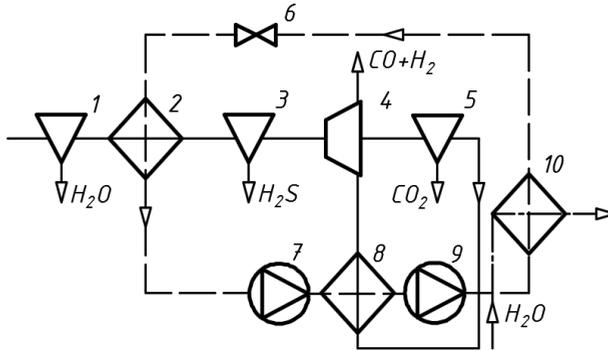


Fig. 1. The principal diagram of low-temperature purification of synthesis gas. The solid line is synthesis gas, the dashed line is the refrigerant cycle, the dash-dot line is water. 1, 3, 5 - cyclone separators; 2, 8, 10 - recuperative heat exchangers; 4 - turboexpander with a compressor on one shaft; 6 - a throttle washer; 7, 9 - 1st and 2nd compression stages.

The calculation is carried out by the thermal balance method. Synthesis gas at the inlet to the unit is wet, the temperature is 323 K, and the pressure is 101.3 kPa, without mechanical impurities. Dehumidification is carried out in the cyclone separator 1. The gas is cooled to 203 K in the heat exchanger 2, i.e., 10K below the condensation temperature of H_2S at atmospheric pressure. As a result, other sulfur compounds such as SO_x are condensing in case of their availability [4]. To purify sulfur compounds in separator 3, the gas must be cooled to 185 K. At this temperature and atmospheric pressure, CO_2 passes from the gas phase directly to the solid state, bypassing the liquid state, as a result of that the so-called "dry ice" is formed (Fig. 2).

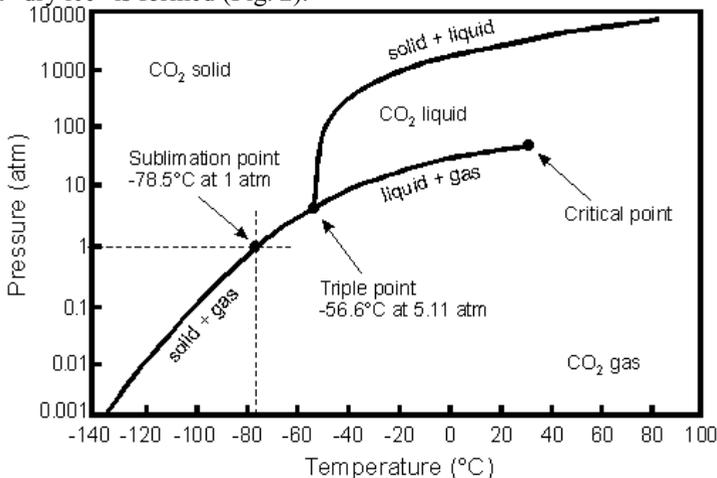


Fig. 2. Pressure-Temperature phase diagram for CO_2 .

A turbo expander 4 must be used to achieve a temperature of 185 K. The synthesis gas purified in a separator 5 is compressed in a compressor and sent to a Fischer-Tropsch reactor.

A refrigeration cycle is used for the heat removal in the heat exchanger 2. It includes two-stage compression 7 and 9, an intercooler 8, a condenser 10 and a throttle plate 6. It is proposed to use R407a as a refrigerant.

4 Results and discussion

The synthesis gas flow rates are taken discretely equal to 5, 50, 500 and 5000 l/min, at a pressure of 101.3 kPa. The heat calculation of the heat exchanger 2 and the calculation of the refrigeration cycle were made according to the scheme (Fig. 1), the corresponding results are summarized in Table. 1.

Table 1. Dependence of performance indicators on increase in the synthesis gas consumption.

The synthesis gas flow rate, l/min	The amount of heat removed from the synthesis gas, kW	Heat which can be utilized usefully, kW	Power consumption, kW
5	0.0244	0.05063	0.0266
50	0.2439	0.5063	0.266
500	2.4391	5.063	2.66
5000	24.3915	50.63	26.6

It can be seen from the Table that 24.4 kW of heat should be removed for the separation of sulfur compounds from synthesis gas, at a flow rate of 5000 l/min, with a cooling coefficient of 1.09. It is necessary to use lower temperatures to remove CO₂ that can be achieved in a turbo-expander. In addition, the consumed power of the refrigerant compressors is 26.6 kW, and it is necessary to raise the pressure of the synthesis gas feeding into the reactor to 2-3 MPa. It is proposed to carry out the purification process at an elevated pressure corresponding to the Fischer-Tropsch reaction process. The parameters of the gas purification process are presented in Table 2, while the schematic of the purification process is shown in Fig. 3.

Table 2. Phase transition temperatures depending on pressure.

Pressure, MPa	Condensation temperature, K	
	Carbon dioxide – CO ₂	Sulfur hydride – H ₂ S
0.1	195 (desublimation)	212.7
2	254	298
3	268	315.3

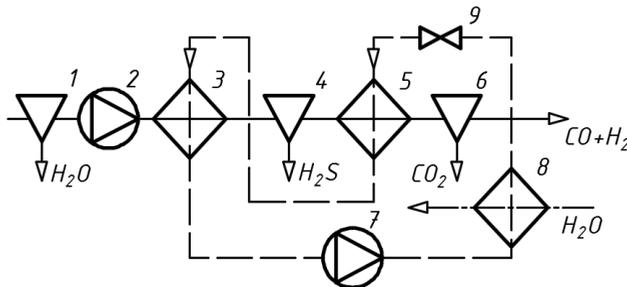


Fig. 3. The principal diagram of low-temperature purification of synthesis gas. The solid line is synthesis gas, the dashed line is the refrigerant cycle, the dash-dot line is water. 1, 4, 6 - cyclone separators; 2, 7 - compressors; 3, 5, 8 - recuperative heat exchangers; 9 - throttle washer.

There is no need to use a turboexpander, since the condensation temperature of CO_2 is above 254 K that significantly reduces the capital costs of the installation. Calculations of the energy costs for the purification of synthesis gas against CO_2 and H_2S , depending on the gas flow rate at different pressures of the cooling process, are shown in Fig. 4, and at a flow rate of 5000 l/min in Table 3.

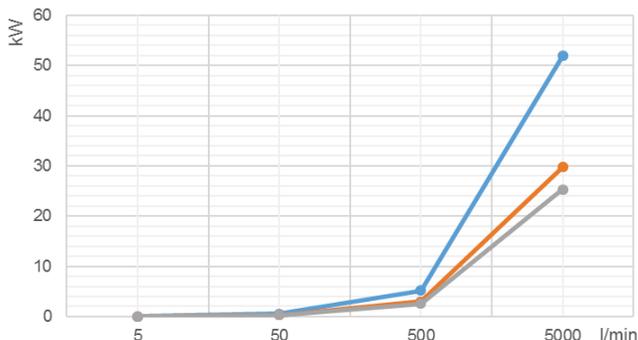


Fig. 4. Dependence of cooling capacity on consumption. Blue line (top) - cooling capacity at 0.1 MPa, orange (in the middle) - at 2 MPa, gray (bottom) - at 3 MPa.

Table 3. Dependence of performance indicators on pressure increase at a consumption of synthesis gas equal to 5000 l/min.

The synthesis gas flow rate, l/min	The amount of heat removed from the synthesis gas, kW	Heat which can be utilized usefully, kW	Power consumption, kW
0.1	51.91	50.63	26.6
2	29.79	48.7	18.9
3	25.34	35.3	9.94

4 Conclusion

Analysis of the calculations shows that the increase in the process pressure and temperature results in decrease in the necessary energy costs for purifying the synthesis gas. The refrigerating coefficient at the pressure of synthesis gas of 2 MPa is 1.58, while it is 2.55 at a pressure of 3 MPa.

References

1. A.R. Bogomolov, S.A. Shevyrev, M.V. Alekseev, *Thermal Power Eng.* **2**, 76 (2013)
2. G. Storch, *Synthesis of hydrocarbons from carbon monoxide and hydrogen*, 257 (1954)
3. A.V. Korobeinikov, *Materials of the II All-Russian Youth Scientific and Practical Conference "Introduction to Energy"*, 23-25 November, Kemerovo (2016)
4. D. Leung, *Renewable and Sustainable Energy Rev.* **39**, 426 (2014)