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Cleaning of Natural Gases from Sour Components

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Abstract. The article presents absorption, adsorption, catalytic and membrane methods of purification of hydrocarbon gases from acidic components, as well as the technology of membrane purification methods.

Keywords: natural gas, acidic components, sulfur, carbon dioxide, absorption, membrane.

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Очистка природных газов от кислых компонентов

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Аннотация. В статье представлены абсорбционные, адсорбционные, каталитические и мембранные методы очистки углеводородных газов от кислых компонентов, а также технология мембранных методов очистки.

Ключевые слова: природный газ, кислые компоненты, сера, углекислый газ, абсорбция, мембрана.

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The capacity of gas processing enterprises in 2014, according to the International Oil and Gas Journal, increased by 30 billion m^3/year (1.1 %) and 2.87 trillion. m^3/year. According to the main indicators of the development of the industry of gas processing plants of Uzbekistan, it shows the dynamics of the growth of the volume of gas extraction and the increase in the coefficient of its use. A large part of the production was increased due to the construction of new facilities, and the rest – due to the expansion of production at existing enterprises [1].

Despite the difficult economic conditions, Uzbek companies continue to dispose of gas processing facilities and associated petroleum gases. In recent years, high rates of LPG disposal have been recorded throughout Uzbekistan, and these rates have been achieved mainly due to the export of liquefied hydrocarbon gases (LPGs).

It is necessary to determine that the complexity of the gas composition affects its preparation and processing. Hydrocarbon gases contain a large amount of sour gas components, water vapor, mechanical particles, salts, a small amount of oil and hydrocarbon condensate [2].

The moisture in the gas has a negative effect on their processing processes, worsens the basic technical and economic indicators (TIK) of the devices and transportation and storage, that is, crystalline hydrates are formed as a result of water entering the condensate pipes. The presence of acidic components in water vapors causes activated corrosion processes. Generally, heavy hydrocarbon gases will have a small amount of water vapor under the same conditions. The presence of hydrogen sulfide (H_2S) and carbon dioxide (CO_2) in the gas composition increases the amount of water vapor, while the presence of nitrogen (N_2) reduces water vapor [3–5].

The inclusion of condensate in the liquid form in the composition of gases complicates the operation of the drying device and low-temperature separation of the gas, the gas has a shock effect on the moving parts of the compressors, that is, as a result of such an effect, it causes them to spread before their time. Thus, one of the most important stages of gas processing is to prepare it for preprocessing.

Cleaning of hydrocarbon gases from acidic components and inert gases and water vapor is carried out using the following methods [6–8]:

- adsorption;
- absorption;
- catalytic methods;
- membrane technology.

Adsorption processes are a cleaning method based on the absorption of acidic components in solid absorbers and are carried out by chemical and physical methods. The main difference between these two types of adsorption is the energy difference of bonds [6].

Absorption is based on chemical bonds of sour gases and sulfur compounds – chemisorption, or solubility of sour components – physical absorption and their combination (joint effect) [7].

In physical adsorption, there is no change in the electronic structure of atoms or molecules. Physical adsorption caused by Vander-Waals forces is the interactions between absorbate and adsorbent molecules. These forces are not so great that there are no active barriers. For physical adsorption, the reverse process (adsorbent regeneration) and multilayer adsorption are suitable. In addition, the process is carried out only at relatively low temperatures [8].

Physical adsorption can be carried out on activated carbon, silica gels and aluminum gels [8]. However, due to the small capacity of adsorption, the effectiveness of heavy hydrocarbons is low, so synthetic zeolites are used in industrial cleaning. They are characterized by their ability to select polar molecules and have a large absorption capacity. CO_2 and H_2S are effectively absorbed by CaA, NaX and NaA branded molecular nets. Desorption stages are carried out by heating adsorbents, vacuuming, spraying inert gases and require large energy costs.

Chemical adsorption or chemisorption has chemical bonds occurring between adsorbate and adsorbent, the heat of chemisorption is ≈ 100–400 kDj/mol. Molecules of adsorbate and adsorbents should have such an energy, which should be higher than the value of the activation energy at the entrance [8].

Among the chemical methods, iron and zinc oxides are widely used in industry. However, the data shows that due to the low technology of the processes, the high degree of non-regenerability and the need to dispose of the used sorbents, they are rarely used [9].

Therefore, the prospect and cheapness of using methanol and water as an absorbent is related to its low enough cost, on the other hand, it has a number of disadvantages, such as the fact that it has very low carbon dioxide absorption properties and not high selectivity.

In recent years, the «Retizol» process has been used for the purification of natural gases from heavy hydrocarbons with a low content, which is based on the absorption of CO_2 and H_2S with cold methanol (–60 ÷ –70 °C) [10]. «Fluor» process is used with the help of ethylene carbomite when the natural gas contains high CO_2 and low H_2S/CO ratios [11].

«Seleksol» process is widely used among physical processes in gas purification [10]. Celexol – dimethyl polyethylene glycol ether is widely used here as an absorbent. Selexol's advantages include: removal of all sour components and all « sulfur organics», targeting of H_2S in the presence of CO_2, inactivity to corrosion. But its use is limited in the presence of heavy hydrocarbons. It can only be used for cleaning dry gases to remove heavy hydrocarbons. Physical absorbents are widely used in «Purizol» processes, based on N-methylpyrrolidone, «Estasolvat» – tributyl phosphate.

In chemisorption, acidic gases react with the active components of the absorbent, causing the formation of chemical compounds, which, when the temperature rises, fall into the primary components.

From chemical absorbents, alkanolamines are widely used on an industrial scale: when amines react with sour gas components, they form sulfides (hydrosulfides and carbonates) and bicarbonates.

Among the chemical sorbents, ethanolamine cleaning method is most widely used in gas processing. In most cases, monoethanolamine (MEA) and diethanolamine (DEA) are widely used as absorbents. These absorbents are replaced by methyldiethanolamine. Imidazole is added to the composition of MDEA in order to carry out the process efficiently [10].

A concentration of up to 40 % of an aqueous solution of diisopropanolamine (DIPA) is used as a chemosorbent. DIPA provides gas separation from H_2S to – 1.5 mg/m^3 and CO_2 to – 200 mg/m^3 (up to 0.01 %) with low solubility in hydrocarbons. In this, up to 50 % of COS and RSH are removed. DIPA forms easily regenerated compounds with CO_2, COS and RSH. The loss of DIPA during the regeneration period is two times less than that of MEA.

Experiments using these processes allowed us to clarify the shortcomings and advantages of each compound (Table 1). Common disadvantages of these processes are as follows [10]:

- high energy consumption (about 70 %) in absorbent regeneration and heat removal;
- corrosion activity of alkanamines.

Table 1. Comparative characteristics of amine absorbents

Purification method in monoethanolamine	
Advantagelari	Very high purification from CO_2 and H_2S; UVs are poorly absorbed; High reactivity; Low price and availability.
Disadvantagelari	Large loss in evaporation; Low efficiency in cleaning from mercaptins; of targeting H_2S in the presence of CO_2; Low saturation of the solution.
Cleaning method in diethylamine	
Advantagelari	High saturation; Stability to the chemical process; Easy regeneration; Excellent gas purification from CO_2 and H_2S in the presence of COS and CS_2.
Disadvantagelari	Absorption property is low; The height of the price; High consumption of absorbents and high energy costs; Low removal of mercaptins and h. CO_2 with non-regenerative compounds.
Purification method in diisopropanolamine	
Advantages	$CO_{2,}$ H_2S, COS, RSR from the surrounding gas is low; Formation of easily regenerated compounds; The width of the range of working parameters; Targeting of the ratio to H_2S in the presence of CO_2; Does not cause a corrosive environment.

In order to solve these problems, inhibitors are added to the solution of ethanolamines based on the protection of the limit of corrosion, that is, it is possible to reduce the circulation of the absorbent and reduce the energy cost of recycling, increase the performance of the device, and reduce the rate of corrosion of the equipment.

The admissible absorption capacity of the absorbent is limited by the admissible corrosion of the apparatus and the marginal admissible chemisorption. Gas purification is carried out in the «Ekonamine» process, which uses diglycolamine solution (DGA) as an absorbent. The use of DGA instead of MEA provides an opportunity to reduce the consumption of absorbent and heat energy costs, but its disadvantage is the high solubility of propane [11].

When the gas contains a large amount of CO_2 and H_2S, gas purification is carried out with the help of diethylene glycol (DEG) and triethylene glycol (TEG), which simplifies the purification technology here, as well as being absorbed by sour components and water vapor [11].

CO_2 and H_2S to acidic components are characterized by a high level of selectivity and a cleaning index.

When alkaline solutions are used, a high level of gas purification from sulfur-containing organic compounds is achieved. This method is used when the amount of H_2S and CO_2 in the gas is not large. For this purpose, alkaline solutions (KON, NaON), arsenic, and alkaline absorbers are used.

One of the first processes for the separation of sulfur compounds is purification using a solution of iron hydroxide. But the formed iron sulfide (Fe_2S) is difficult to regenerate and increases corrosion.

Today, the process is technologically improved and it is possible to obtain pure sulfur as a commodity product.

Physico-chemical absorption processes use a mixture of physical absorbent with chemical absorbent, which is called combined absorbents. The different action properties of each absorbent make it possible to clean the gas not only from sulfur and carbon dioxide, but also from sulfur compounds. Currently, the most widely used combined absorbents in industry include sulfanol, which is a mixture of diisopropanolamine (30–40 %), sulfanol (tetrahydrothiophene dioxide 40–60 %) and water (5–15 %). Shell has proposed a technologically improved process by coupling with Sulfanol's SSOT unit [12].

from hydrogen sulfide and organic compounds of sulfur, CO_2 "Ukarsol" absorbents, which provide the possibility of cleaning, are widely used.

In addition, in addition to the indicated methods for cleaning gas from acidic components, there are also catalytic methods that are based on the oxidation and recovery of acidic gases with the participation of nickel, cobalt and other catalysts. This method is used when there are compounds in the gas, when there is no possibility of complete removal using liquid absorbers or absorbents (sulfur, carbon dioxide of sulfur, sulfides, disulfides, thiophenes).

When reduction reactions are carried out under the influence of hydrogen (hydrogenation) or water vapor (hydrolysis) using aluminum-based catalysts of cobalt oxide, nickel, molybdenum, primary compounds precipitate into hydrogen sulfide and compounds, the precipitate does not contain sulfur.

Oxidation methods are widely used in industry, and hydrogen sulfide is reduced to elemental sulfur or mercaptides to disulfates (Merox process) in activated aluminum oxide. The achievement of the process is selectivity when there is no need to extract CO_2. A secondary disadvantage of conducting the reaction is that it increases the consumption of reagents, deposits appear on the walls of the equipment and cause a corrosive environment.

The following processes are also known based on the oxidation method:

- Use of ammonia or soda solutions and hydroquinone catalysts as «perox» absorbers;
- «Townsend», where DEG is used together with dissolved sulfuric anhydride;
- «Haynes», regeneration of H_2S accumulated in membranes with the formation of sulfur under the influence of boiling SO_2;
- «Ferox», using an aqueous-alkaline solution of iron hydroxide and subsequent regeneration of $FeS(OH)_3$ and sulfur formed in the product;
- «Stranford», an aqueous-alkaline solution of anthraquinone-disulfuric acid and salt vanadium is used;
- «Lo-Sat» shows the ability to dissolve iron in water when iron catalyst is used as a reagent and ethylenediaminetetraacetic acid is used as an agent [13].

There are combined methods in gas purification, where the catalytic method is combined with the absorption method. SSOT is one of the advanced processes for separating gas sulfur compounds [13].

This defined technological process is divided into three sections:

1. In the regeneration reactor, all sulfur compounds contained in the gas are transferred to hydrogen sulfide.

2. Rapid cooling section, the gas leaving the reactor is cooled, and the water is condensed (turns into a liquid).

3. In the absorption section, H_2S is selectively absorbed in the amine solution. The charged solution is regenerated, and the discharged sour gas is returned to the inlet of the device.

The Shell Slaus off-gas plant is easily compliant with environmental regulations under strict sulfur emissions and carbon monoxide emissions limitations.

One of the most promising areas of gas purification is the use of membrane technology.

have been applied to industrial production in CO_2 separation processes, and they have been used only in the treatment of small flows of natural gas. The share of membrane technology in the market is increasing due to the separation efficiency of the membrane system [14].

CO_2 from natural gas are made of cellulose acetate or triacetate.

The efficiency of using membranes is characterized by two key parameters: permeability and selectivity.

The improvement of membranes is being carried out due to these indicators: when the permeability is increased, the area (surface) of the membrane necessary to ensure a clear separation is reduced; selectivity is increased in order to increase the degree of purification of the product.

To determine the effect of these parameters, we consider the comparison of the permeability of clean gas through two sample membrane materials used in natural gas purification: silicone rubber (SR) [14] and cellulose acetate (SA) [15, 16].

The SR polymer has flexible polymer chains and its selectivity depends on the very large molecules, which (S_3N_8) propane is condensable to methane (SN_4) and therefore permeable to methane.

SA has rigid polymer chains, its selectivity is often determined by the selectivity diffusion coefficient, the gas permeability of SA is in the following order, looking at Fig. 1 from above: H_2, CH_4, ethane (C_2H_6) and C_3H_8) [16, 17].

H_2O, CO_2 and H_2S compared to SN_4 in both rubbery states of SR and glassy polymers of SA due to the small size of molecules, high diffusion coefficient and high condensation compared to SN_4N_2 O, CO_2 and H_2S is more conductive. Both membranes have been used to remove H_2O, CO_2 and H_2S, which in most cases CO_2 removal is accomplished using glassy polymers of cellulose acetate.

Gas permeability is inversely proportional to the layer thickness of the membrane separator, industrial membranes have very thin selective layers and provide a large flow. In addition, the membranes must have sufficient mechanical integrity to withstand pressure differentials during treatment, where natural gas streams are often treated at high pressures. To solve these problems, it is necessary to provide a very thin layer, but mechanically strong membranes, such as anisotropic coatings, are considered [18]. A layer of thin density material in such membranes provides molecular separation of gases, while the porous mass of the membrane provides mechanical stability, does not show any resistance to mass transfer.

Currently, the membranes used in natural gas purification are produced in the form of flat sheets or hollow tissues. Flat sheets are packed in coiled modules in the form of twists, while hollow tissues are combined into bundles (multi-textured modules), which are in the form of sheathed tubular heat exchangers.

Packaged type devices are in the form of modules packed in a steel package placed in series (Fig. 2).

Gas transfer passes axially to the module through the membrane coating. Highly conductive components like H_2O, CO_2 and H_2S passes through the membrane, enters the center and is removed through the collection tube.

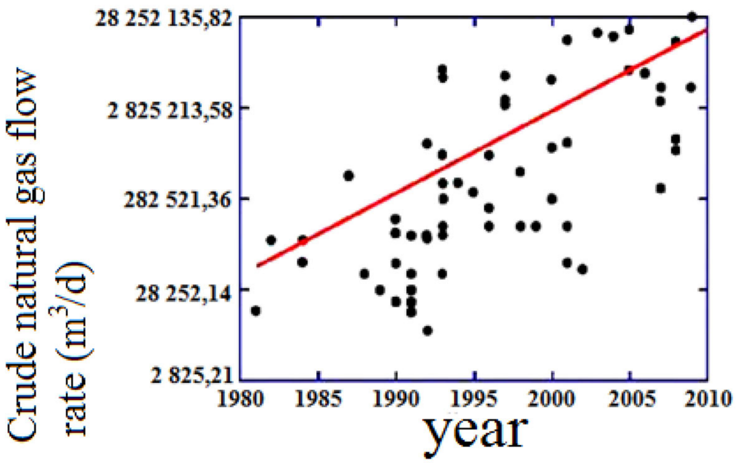

Fig. 1. Performance indicators of a membrane device for cleaning gas from sour components

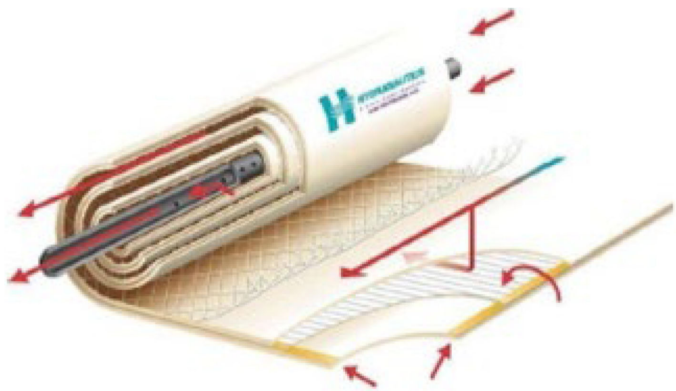

Fig. 2. Packaged module

The hollow tissue module (Fig. 3) consists of a bundle of hollow tissues, the diameters of which are several tens and hundredths of a millimeter, and are hermetically fixed by themselves to the body of a cylindrical shape. Gas transfer takes place through the interstices of tissues. The passed gas moves through the tissue until it reaches the permeate in the pores.

Currently, both types of membrane modules are produced for natural gas purification. He has not identified a clear leader in the market. Multi-texture modules allow large surface area membranes to be placed in compact membrane a modules. In high-pressure processes, the diameter of the tissue is reduced and the thickness of its wall is increased to increase the mechanical strength of the tissue.

The disadvantage is a decrease in performance due to an increase in the pressure difference of the permeate when the inner diameter of the tissue is reduced. When the thickness of the hollow tissue wall is increased, the total area of the outermost membrane increases.

In practice, under high pressure conditions, the permeability of a flat-sheet membrane is high when it is formed in the form of a twist-wound module. This situation can compensate for its higher cost compared to hollow tissue modules [19].

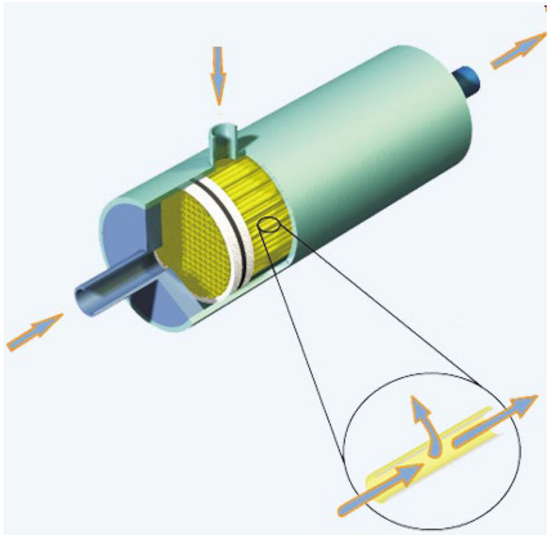

Fig. 3. Multi-tissue module

Membrane systems are often compact – occupying a small area, slow – when the parts do not move, reliable – there is no need for constant attention to work in remote areas. Membrane technology separates gas based on the pressure of forces moving through the membranes. Here it is necessary to take into account the risk of factors under the influence of high pressure.

The actual buoyancy force is similar to the buoyancy-based buoyancy force. As for a pollutant such as CO_2 and normal butane (n-C_4H_{10}), the actual kinetic energy based on volatility is much lower than the kinetic energy under pressure. For SO_2 and n-S_4N_{10}, the actual gas flow will be very small compared to using the predetermined gas partial pressure. If this effect is not taken into account in the design of membrane systems for CO_2 removal, then the expected separation efficiency cannot be achieved.

Gas permeability and selectivity are strongly affected by the decrease in operating temperature. Often, membranes based on glassy polymers have a very low permeability for higher hydrocarbons compared to methane, that is, such a situation leads to a constant enrichment of higher hydrocarbons in the permeate. An increase in the content of higher hydrocarbons increases the dew point of the gas, which, when combined with a decrease in temperature, can cause condensate to form in the precipitate. Condensate on the surface of the membrane reduces gas permeability and can damage the membrane, shortening the service life of membrane modules. A reliable membrane system must be designed in such a way that it is necessary to avoid the possibility of hydrocarbon condensate in the modules.

Pollutants such as CO_2 and heavy hydrocarbons in natural gas can be adsorbed in membrane polymers to a large extent. An experiment was conducted to determine the effect of the composition of natural gas on membrane materials, which is presented in [20]. The crude natural gas was collected from several wells and contained a large variety of hydrocarbons and hydrocarbon compositions. The modulus of the gas solubility ratio with the critical temperature of the gas was used. The results show that the polymer CO_2 ~3.6 % of mass and ~8.9 % of hydrocarbons can swallow mas sas, Later, when their concentration is increased, it causes them to change in size to a large value and has the ability to separate gas.

The high pressure conditions of natural gas processing provide high driving forces for membrane processes and present several unique challenges, namely gas phase nonideality, plasticization of membrane materials, and the possibility of condensation of heavy hydrocarbons.

The use of membrane technology in sour gas component processes reduces operating costs in practice. However, this does not completely solve the problem of gas purification. When a very high level of purification is required, which membrane technology alone cannot provide, combined purification schemes are often used.

Sour gas SO_2 The hybrid purification scheme from [21] is shown in Fig. 4. The main mass of sour components is separated in the membrane block, and then the gas is directed to the amine block to completely purify the gas stream. Reducing the flow of gas and sour components in the amine treatment unit allows to minimize the absorbent circulation, thereby achieving a significant reduction in energy costs.

With this scheme, it is possible to obtain a product of the same quality as treated with DEG, but with reduced size of the amine treatment plant and lower capital and operating costs.

Currently, the membrane technology does not allow simultaneous separation of sour gas from SO_2 and H_2S, where the permeability coefficient of these components is close. When it is necessary to separate H_2S and CO_2, there are practical options to purify the H_2S upstream (for example with amine) and then remove the CO_2 in a gas membrane scrubber.

Absorption ethanolamine purification is the most widely used technology for hydrocarbon gas purification in modern production. Therefore, when evaluating the economic efficiency of using other technologies, a comparison with the technology carried out in one or other conditions is made with the ethanolamine absorption technology.

When choosing a technology, the following can be considered as parameters affecting it:

- the presence of water vapor and hydrogen sulfide in the raw hydrocarbon gas;
- volatility of absorbent;
- durability of adsorbent;

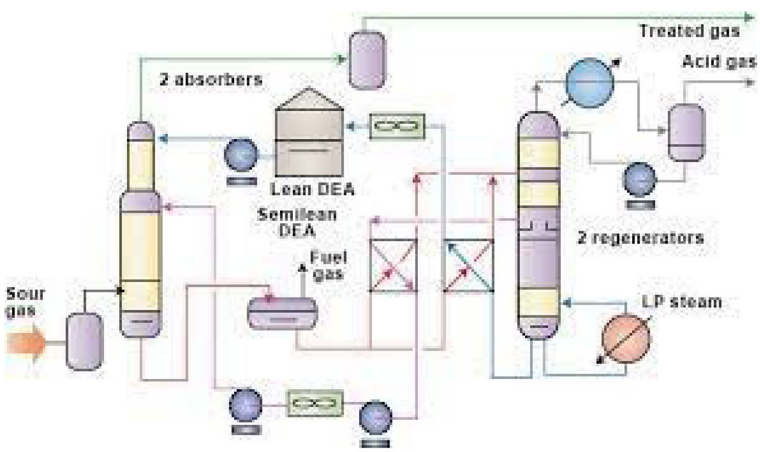

Fig. 4. Hybrid scheme of sour gas purification from sour carbon gas

- selectivity, complexity of regeneration;
- price and service period.
- In addition, in the case of membrane comparison, permeability, consumption and pressure of the raw stream are taken into account.

Technical-economic calculations of the effectiveness of the membrane application, for gas of different composition, absorption or combined technologies for cleaning gas from sour components, and production scales, the following conclusion can be drawn:

- is useful in the purification of gases with a low concentration of SO_2 and H_2S, which are processed in a large volume;
- has an advantage in production processes with a high CO_2 content in the raw stream and relatively small volumes;
- CO_2 is high and the costs of the raw material flow are high, the use of the combined technology is more economically efficient.

Factors of the location of the object are of great importance. Membrane technology has significant appeal in offshore projects due to its small size and low cost in meeting safety requirements.

The advantages of combined schemes of separation are the high degree of smoothness of the process in relation to the composition and the parameters of the raw material flow.

Summary. According to static data, the global volume of gas processing with the help of membranes has reached 5 %, and the largest share is amine purification technology – 62 %, and other technologies – 10 %, and the rest corresponds to the share of untreated gas [22].

Depending on the cleaning index, the most effective methods of cleaning gases from hydrocarbons are selected, taking into account the type and composition of the processed gas, the location of the device, and the provision of its costs. The main factors of the application of one or another type of cleaning technology are determined taking into account the technical and economic indicators.

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