

OBTAINING HIGH-PERFORMANCE COMPOSITE MATERIALS BASED ON NAVBAHOR BENTONITE AND STUDYING THE SORPTION PROPERTIES

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ABSTRACT

Synthetic zeolites based on Navbahor bentonite CaA, NaA, HSZ and NaY were tested in order to select the optimal adsorbent for deep purification of natural gas from various unwanted components. Among the tested zeolites, NaY zeolite is resistant to bitter environments and HSZ based on Navbahor bentonite are effective for zeolite decomposition. The total adsorption capacity of synthetic zeolite obtained on the basis of Navbahor bentonite at a temperature of 25 °C is 182 g/l according to H₂S. A mathematical model of adsorption purification of natural gas was developed, the design parameters of the process were determined. The main purpose of the work is to synthesize highly effective sorbents based on Navbahor bentonite and optimize the process of purification of natural gas and oil products from impurities.

Keywords: synthesis of highly effective sorbents, zeolites, Navbahor bentonite, natural gas and petroleum products.

INTRODUCTION

Natural gas is a gaseous fuel compound, the main component of which is a low molecular weight hydrocarbon (alkanes C₁ - C₄). It contains partly hydrogen carbon monoxide and hydrogen sulphide [1-4]. In the process of obtaining natural gas, it contains moisture, and at low temperatures, they form hydro crystals with gas molecules. The amount of water in the contents accelerates the corrosion process of the devices, resulting in accidents, which lead to rapid failure of gas pipelines gas processing plants. Therefore,

the loss of moisture and water in the natural gas must be purified for the transmission and processing of natural gas, the moisture in the gas is determined by the dew point temperature and controlled within the allowable norm (network standards). Drying is the most important process in preparing natural gas for transmission in main gas pipelines. All gases transported from the main pipelines must be dehumidified and must meet industry standards, technical specifications must be suitable for further processing [4-5]. The drying process of natural gas is mainly carried out in four industrial methods: by cooling, by the absorption method, by adsorption method and by a combination of three methods. In addition, the known methods of gas drying are divided into three groups according to the classification sign: physical, chemical, physicochemical [6-8]. The most widely used method in industry is the bifunctional absorbent method, which uses three groups of absorbents: diethylene glycol and triethylene glycol monoethyl ether; N methyl pyrrolidone (N MP); solar oil mixture of diethylene glycol. The increase in the performance of the gas purification process in a zeolite (lowering the dew point in moisture, increasing the purification from sulfur) is sufficiently improved, in which it is formed in the form of three papers in zeolites of type 4A, 4A and 5A. The use of two-layer adsorbents in the adsorption system to improve the drying and purification process of gases gives good results. One layer consists of zeolite balls (granules) of different shapes and diameters, the second layer consists of a front layer in the form of aluminium oxide [2-6].

At present, due to the important properties of bentonite clay, a lot of attention is paid to it, mainly due to its adsorption properties. The bentonite industry is widely used in various fields, currently used in more than two hundred areas of production, including the treatment and preparation of hydrocarbon gases, but there is little information in the literature, including in the process of natural gas purification. The low cost of bentonite sludge has had a significant impact on its cost in addition to production, one kilogram of bentonite powder is much cheaper than other types, and its production, processing and sale are much cheaper than petroleum products [8-9].

From the above, it should be noted that the use of bentonite sludge and zeolites as adsorbents is currently important for the drying, purification and preparation of natural gas. 98.5% ethylene glycol was obtained as an adsorbent. A special device has been developed for laboratory tests, in which the ability of adsorbents to absorb moisture and other (bitter) components is determined. The absorbency of reagents is assessed at the dew point temperature in natural gas [3-8].

MATERIALS AND METHODS

Adsorption experiments were performed on a laboratory device in the following way.

The adsorbent coating was activated in a muffle furnace at a temperature of 784K for 2.5 h and then cooled in a desiccator to constant weight (to the nearest 0.001g). The natural gas purification process was studied in a sampled device at different temperatures in a sample adsorbent. Natural gas was transferred from a special cylinder to the adsorption column (adsorber), which is in the form of a cylinder with a total height of 210 mm, a diameter of 45 mm (volume 8.45 cm³). The adsorber was mounted in the furnace with adsorbent. The adsorption temperature was monitored using a thermocouple mounted on the adsorber. The stove was heated on 220V mains. The gas passing through the adsorbent layer falls into a gas meter filled with brine (saltwater), at which time the saltwater is compressed in a cylinder. The pressure in the adsorber is equal to atmospheric pressure and it is monitored using a special glass monometer using a saturated solution of NaCl in the gasomer. The amount of water and acidic components (H₂S and CO₂) from the second column is determined after the first column before the purified gas is passed to the chromatograph connected to the gas analyzer by a special device after the gas meter. The experiments were performed at given temperatures under atmospheric pressure. The raw material was sent for 40 minutes, 50 g of adsorbent was added. Sample adsorbents in the device were tested for adsorption processes at temperatures of 20, 25, 40, 45, 40 °C. Prior

to the experiment, the adsorbent was activated by nitrogen spraying for 10 min. Based on the results of the analysis, the adsorbent dynamic activity of bentonite HSZ obtained from Navbahor [10-15] was determined using the following formula.

$$A_0 = C_0 \cdot W \cdot t \cdot h$$

In this case, the initial concentration of H₂S, CO₂ and NO₂ in the S0-mixture, g/100g; W – gas flow rate, m/s; τ-adsorbent protective exposure time; h is the height of the adsorbent layer, m. We studied the adsorption of CO₂, H₂S, and NO₂ isotherms on an activated carbon adsorbent of SKT and HSZ derived from Navbahor bentonite. Adsorption isotherm was determined in HSZ zeolite obtained from Navbahor bentonite [16-28]. HSZ derived from Navbahor bentonite were found to be suitable for the separation of zeolite CO₂, H₂S, and NO₂ adsorption. The amount of adsorption isotherm is written by the Langmuir equation.

RESULTS AND DISCUSSION

Adsorbent samples Adsorption activity was tested at different times of contact with water. Figure 1 shows the results of the moisture capacity testing of these adsorbents obtained in HSZ Navbahor bentonite, NaA and CaA, aluminosilicate and HCl activated with solar acid-activated bentonite and Na₂CO₄ carbonate sodium) adsorption time relationship, natural gas drying process at atmospheric pressure.

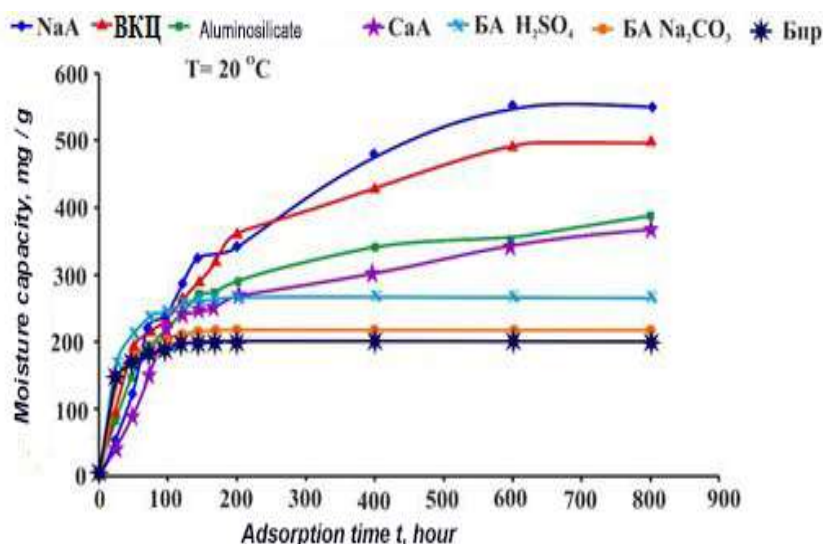


Figure 1. Moisture adsorption isotherm in different adsorbent samples.

We also studied the adsorption and moisture adsorption of activated bentonite sludge in different ways. When carbonate is activated with sodium, an alkaline substance is formed, which affects the chemical composition of bentonite, as a result of melting, the content of silicon oxide in free silicon decreases and aluminium oxide, iron, alkaline earth elements increase in bentonite pattern. In sodium bentocomplexes, when sodium ions are mixed with metals of the first group in the table of elements, they are observed to increase up to four times, which in turn increases the ion exchange of sludge from 85 to 120 mg-eq/100g. When aluminium, iron, and magnesium ions are treated with acid, the crystalline structure of the clay minerals is disrupted and the surface area increases as a result of washing. The bentonite particle size was reduced, more precisely, the structure was improved by reducing the diameter from 118nm to 77nm. The specific surface area of bentonite increases threefold to 87 m²/g. It can also be said that HCl – 178 mg/g activated bentonite has high adsorption, its natural activity is 150 mg/g. This can be explained by the fact that bentonite activated with sulfuric acid has a large specific surface area, 87 m²/g instead of 24 m²/g. The moisture absorption coefficient of the indicated adsorbents was determined by the desiccator method. The

duration of adsorption was 800 hours. The moisture content of the adsorbents was measured first, every 8 days for 8 days, then once every 200 hours. The results of the analysis show that the adsorption isotherm shown in Figure 2 shows that the moisture capacity of the adsorbents depends on the duration of the experiment, while the water vapour adsorption varies depending on the test time.

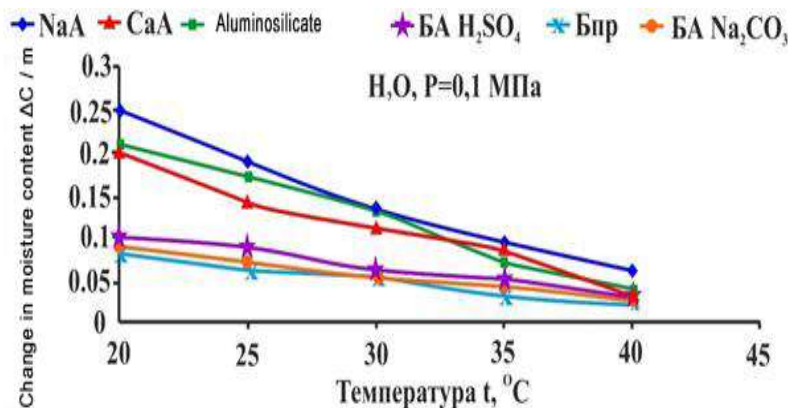


Figure 2. Humidity content changes with temperature.

When the adsorption duration is 20-100 hours, the moisture capacity in bentonite activated with sulfuric (aolnic) acid is large. Its value is in the range of 180-240 mg/time at this time. It is followed by natural bentonite, the moisture absorption of 20 hours of adsorption increases from 145-150 mg/g to 200 mg/g in 5 hours. On the first day of the experiment, the adsorption efficiency is in the following sequence: CaA (40 mg/g); NaA (40 mg/g); aluminosilicate (85 to 80 mg/g); HSZ obtained from Navbahor in bentonite (95 mg/g); in natural bentonite (145 mg/g); In bentonite activated with HCl, (175 mg/g).

On the fourth day of the experiment (duration 100 h), the moisture capacity of the adsorbents was changed in the following sequence: natural bentonite (190 mg/g); bentonite, bentonite activated with Na₂CO₄ (210 mg/g); CaA (220 mg/g); HSZ bentonite from Navbahor (245 mg/g); NaA (245 mg/g); HCl activated bentonite (245 mg/g). Humidity for the next 4 days was as follows: natural bentonite (200 mg/g); Activated bentonite with Na₂CO₄, (220 mg/g); HCl-activated bentonite, (280 mg/g); CaA (280 mg/g); alyumo-silicate (290 mg/g); NaA (450 mg/g); Ventures bentonite (275 mg/g) obtained from Navbahor. Subsequent adsorption time (800 hours) The change in moisture capacity of the adsorbents seems to have stabilized somewhat in the experimental results, and when it is processed for 200 hours: in natural bentonite (200 mg/g): In Na₂CO₄ -activated bentonite (200 mg/g) in HCl-activated bentonite (280 mg/g). Moisture capacity in zeolite and aluminosilicate increases with increasing adsorption time. NaA zeolites and HSZ from Navbahor were stabilized in bentonite after reaching the maximum moisture content for 25 days: NaA - 550 mg/g, Navbahor HSZ from bentonite - 500 mg/g. At 44 days, experiments showed that the moisture content of NaA zeolite decreased slightly. Moisture capacity of aluminosilicate and SaA in zeolite increases by 800 hours Moisture capacity: up to 490 mg/g in aluminosilicate and up to 485 mg/g in SaA at zeolite. Thus, comparing the adsorption isotherms, it can be seen from Figure 2 that a large amount of water is adsorbed on NaA zeolite: moisture content from 400 mg/g to 550 mg/g when processing time increases from 280 to 800 hours, in Navbahor HSZ bentonite moisture content from 400 in the same time interval. Increased to 500 mg/g. The most effective in drying natural gas is NaA zeolite and HSZ bentonite from Navbahor, which have increased moisture absorption per 100 hours and increased the adsorption process. When the adsorption process is up to 100 hours, the best adsorbent is SaA zeolite. Adsorbents derived from bentonite sludge were saturated during the initial absorption adsorption (20–100 h) and then maintained stability. Figure 2 shows the adsorption activity of adsorbents in water (zeolites, aluminosilicates and bentonites) at

different temperatures. The effect of temperature on atmospheric pressure was studied in the temperature range of 20–40 °C. Adsorbent samples were studied as a measure of adsorption relative to the amount of water in natural gas samples relative to the mass of the adsorbent ($\Delta C/m$). The moisture content of natural gas samples was studied chromatographically. As the temperature increased, the adsorption activity of adsorbents relative to water decreased. NaA zeolite has adsorption activity on water vapour. The change in moisture absorbed at a temperature of 20 °C ($\Delta C/m$) is 0.25 instead of 0.2 in aluminosilicate, 0.19 in SaA zeolite, 0.1 in bentonite, 0.09 in HCL: 0.09 in bentonite, 0.085 in Na_2CO_4 : 0.085 in natural bentonite. When the temperature rose to 40 °C, the moisture content in zeolite was 0.08 NaA, 0.04 in aluminosilicate, 0.04 in SaA, and 0.025 in zeolites (natural, activated with sulfuric acid and sodium carbonate). The absorption capacity of an adsorbent during an experiment is called its working or dynamic activity. Dynamic activity is always lower than static activity and it is related to the operating conditions of the adsorbent. The dynamic activity of the adsorbent is a key indicator in determining the size of the adsorbers and in determining the time of the sorption cycle. Zeolites are mainly used to purify the gas from H_2S , CO_2 and NO_2 . The characteristics of sorbents (HSZ zeolite sample from Navbahor bentonite) are given in Table 1.

Table 1. Adsorption properties of Navbahorskogo bentonite

| Indicator | Example | | |
|--|---------|-------|-------|
| | 1 | 2 | 4 |
| Density, g/cm^4 | 0,885 | 0,880 | 0,848 |
| tumour (v mm),% | | | |
| 2,8-2,0 | 44,2 | 44,0 | 41,7 |
| 2,0-1,5 | 74,8 | 72,8 | 74,8 |
| 1,5-1,0 | 4,1 | 4,2 | 4,7 |
| Contents, % | | | |
| Zol | 8,4 | 8,1 | 8,9 |
| potassium | 0,20 | 0,27 | 0,28 |
| Sulfur | 0,47 | 0,48 | 0,78 |
| Particle size, cm_4/g | | | |
| V_{MH} | 0,28 | 0,42 | 0,44 |
| V_{Mc} | 0,11 | 0,14 | 0,12 |
| V_{Ma} | 0,28 | 0,41 | 0,45 |
| V_{Σ} | 0,78 | 0,87 | 0,80 |
| Structural constants $W_0 \text{ cm}^4/\text{g}$ | 0,29 | 0,44 | 0,45 |
| V 107 (for benzene) | 0,44 | 0,54 | 0,54 |

The adsorption process allows the complete removal of unnecessary components from the gas mixture and deep cleaning of the gas. Therefore, this method is more effective than other methods in protecting the environment and quality gas cleaning. Adsorbents used in the purification of effluents must meet the following requirements: high adsorption capacity when absorbing components, low concentration in gas mixtures, high selectivity, and high mechanical strength, low cost and good regenerative properties.

Table 2. Static and dynamic activity values of HSZ and SKT activated carbon adsorbents derived from Navbahor bentonite

| Adsorbents components | SKT Coal Active. | | | Bentonite HSZ from Navbahor | | |
|--|----------------------|---------------|---------------|-----------------------------|---------------|---------------|
| | H_2S | CO_2 | NO_2 | H_2S | CO_2 | NO_2 |
| Statistical activity, $\text{g}/100 \text{ g}$ | 9,88 | 8,74 | 7,97 | 12,28 | 10,51 | 9,85 |
| Dynamic activity, $\text{g}/100 \text{ g}$ | 8,02 | 5,24 | 4,84 | 9,89 | 8,44 | 8,81 |

When analyzing various adsorbents for the adsorption of CO₂, H₂S and NO₂ from natural gas, it is advisable to use HSZ derived from Navbahor bentonite. Adsorption isotherms, as well as dynamic activity, are important in computer modelling. The dynamic activity of adsorbents is important in determining the adsorber size and sorption cycle time. HSZ zeolite obtained from Navbahor bentonite, a gas mixture of CO₂, H₂S and NO₂, was passed through the top of the adsorber at a pressure of 1-7MPa and a temperature of 20-400 °C. Adsorption of CO₂, H₂S and NO₂ occurs in the stationary part of HSZ zeolite obtained from Navbahor bentonite. The process is carried out in 4 adsorption devices. The first adsorber is in the adsorption process, the second in the desorption, the third in the regeneration, and the fourth in the cooling. The natural gas flow rate is determined from the hydraulic resistance of the adsorbent layer. The zeolite layer of HSZ derived from Navbahor bentonite is saturated with unnecessary components of CO₂, H₂S and NO₂. Once the zeolite is fully saturated, the adsorber goes directly into the regeneration mode and the process continues like this. Natural gas consumption is controlled by a special meter (flowmeter), the humidity of the gas leaving the adsorber is measured using the instrument "Parametric-280". The concentration of the components of the gas mixtures is determined in the Rubotherm instrument. The decrease in the pressure of the adsorbent layers to different values allows determining the concentration of gas mixtures. Depending on the adsorption process, the gas concentration value and the exit time of the purified adsorber are given in Table 3.

Table 3. Values of the concentration of gases in the purified stream at the outlet of the adsorber depending on the conditions of the adsorption process

| The values of the pressure drop in the adsorption layer are kg/cm ² | Composition of gas mixtures after cleaning,% tar. | | |
|--|---|-----------------|-----------------|
| | H ₂ S | CO ₂ | NO ₂ |
| 0,154 | 0,059 | 0,041 | 0,088 |
| 0,174 | 0,054 | 0,044 | 0,08 |
| 0,184 | 0,048 | 0,018 | 0,08 |
| 0,184 | 0,042 | 0,012 | 0,072 |
| 0,194 | 0,038 | 0,009 | 0,058 |
| 0,204 | 0,041 | 0,010 | 0,078 |
| 0,214 | 0,042 | 0,014 | 0,084 |
| 0,224 | 0,044 | 0,020 | 0,087 |
| 0,244 | 0,048 | 0,027 | 0,092 |

Thus, when using HSZ zeolite derived from Navbahor bentonite, three-component adsorption in a gas mixture by the proposed method at an adsorption layer pressure of 0.184 ÷ 0.204 kg/cm² is H₂S: NO₂:CO₂,% per volume unit - H₂S-80%, CO₂-15%, NO₂- 5%. The optimal option for the adsorption process is calculated when the pressure drop in the adsorption layer is 0.194 kg/cm². In this case, the concentration of components after purification in%: H₂S-0.038, CO₂-0.009, NO₂-0.058. The results of the experiment we obtained show that the gas flow rate is 1.5 m/s and the pressure drop in the adsorption layer is 0.184 -0.204 kg/cm⁴, the unused adsorption volume is approximately 0. When the flow of gases containing carbon dioxide, nitrogen dioxide and sulfur is in contact with HSZ zeolite obtained from Navbahor bentonite, the gas pressure in the adsorption layer is 0.184 ÷ 0.204 kg/cm². The main task of the method is to simultaneously create a method of purification of hydrogen sulfide, sulfur dioxide and NO₂ in the gas. The task is to clean the gas stream in contact with the adsorbent HSZ CO₂, H₂S and NO₂ obtained from Navbahor bentonite at the pressure of the adsorbent layer 0.184 ÷ 0.204 kg/cm². This method allows purifying when the adsorption layer pressure decreases when in contact with HSZ zeolite obtained from Navbahor bentonite in the presence of three-component H₂S: NO₂: SO₂ mixture in%: H₂S-80%, CO₂-15%, NO₂-5%. In addition to adsorption and

catalyst purification methods, the adsorption process is also widely used in the quality purification of natural gas from CO₂, N₂S and NO₂. Adsorption of CO₂, N₂S and NO₂ from gas mixtures is carried out by means of zeolites, silica gel and activated carbon. As a result of experimental studies, the composition of gaseous compounds CO₂, N₂S and NO₂ adsorption isotherms in different adsorbents: HSZ derived from Navbahor bentonite were detected in zeolite, TA 95, TA 120 activated carbon and KSM silica gel. CO₂, N₂S and NO₂ from the gas composition were extracted from different adsorbents and carried out under the same conditions.

The sorption separation of CO₂, N₂S and NO₂ in HSZ zeolite derived from Navbahor bentonite has been shown to be superior to other methods. Boundary static activity of HSZ adsorbent from Navbahor bentonite: CO₂–11.0; N₂S – 12.7 and NO₂–9.8 g/100 g were detected. Purification of gas mixtures from zeolite is a cyclic process. The organization of the process of purification of natural gas with zeolites has certain technological and design difficulties.

We have created a new technology for cleaning natural gas with synthetic zeolites. It was found that the most important disadvantage of adsorbents is that their activity decreases during the sorption process, especially when working with multi-component mixtures.

As a result, in our case, the particle size (granule), working layer height, amount of zeolite adsorbent, temperature regime during adsorption and regeneration are important for natural gas adsorption. During the adsorption process, the rate of natural gas flow through the adsorbent layer was significantly affected. It plays an important role in layer pressure and other exchange processes. We determined the optimal value of the pressure drop in the adsorbent layer. The guaranteed complete absorption of CO₂, N₂S and NO₂ is observed at a pressure limit of 18 p of 18.78 kPa÷19.81 kPa. In this regard, we carried out the adsorption of CO₂, N₂S and NO₂ from natural gas in the artificial zeolite. The result is natural gas isotherm and kinetic adsorption. The design and technical parameters of the process were also determined, the adsorption isotherm corresponds to the type of Langmuir isotherm. As the mixture passes through the adsorbent-filled adsorber, the concentration front (flow) of each component becomes larger. The separation of substances is due to the difference in the retention of the mixture in the stationary phase. Flow washing and motion characteristic adsorption isotherm and non-ideal factors: the equilibrium of the velocity of substances in the moving and inactive phases is established (kinetic braking).

Experiments on the adsorption isotherm and kinetics of NaA synthetic zeolite have shown that complete absorption of CO₂, N₂S and NO₂ is ensured when the adsorbent layer thickness is 5.48 m, the adsorber diameter is 4.79 m, and the adsorbent working layer is 142 cm. It should be noted that if the technological mode schemes and equipment are chosen correctly, H₂S hydrogen sulfide, CO₂ carbon dioxide and NO₂-nitrogen oxide are completely removed from the natural gas from various deposits by adsorption. The new approach not only protects the biosphere from various gases, but also protects devices from corrosion, poisoning from catalysts used in chemical technologies.

CONCLUSION

1. Solid adsorbents - zeolites, cystolite storage catalysts, aluminosilicates, silica gel and bentonite (natural, sodium carbonate and HCl and hydrochloric acid Na₂CO₄ from Navbahor deposit of the Republic of Uzbekistan) to create a promising method for the preparation of natural gas for processing in a methane-hydrogen mixture) adsorption capacity was studied.

2. When studying the adsorption activity of adsorbent samples on H₂O, it was found that NaA zeolite is effective in drying natural gas.

3. It was found that when bentonite sludge is activated with Na_2CO_4 and HCl, their adsorption activity increases. Experimental results have shown that bentonite activity is high in the adsorbents we studied, especially in the initial stage of adsorption on moisture absorption.
4. Although the adsorption activity of natural bentonite sludges is low, we believe that they are promising for future drying because the activation of aqueous solutions of Na_2CO_4 and HCl helps to increase the adsorption activity of bentonite adsorbents.
5. A mathematical model of adsorption purification of natural gases has been developed. The constructive parameters of the process were determined.
6. The diffusion coefficient at repeated values of the heights of the adsorbent layers was checked. As the layer thickness increases, the value of the diffusion coefficient increases significantly. The values of the diffusion coefficient increase in the following sequence by components: NO_2 , CO_2 , H_2S . The largest diffusion coefficient is H_2S , the smallest is NO_2 . Depending on the height of the adsorber also changes the technological parameters that determine the optimal mode of deep purification of natural gas. Therefore, the diffusion coefficient should be taken into account when creating an industrial-based computer model for the deep purification of natural gas.

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