

ADSORPTION SYSTEMS OF NATURAL GAS STORAGE AND TRANSPORTATION AT LOW PRESSURES AND TEMPERATURES

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Questions of the development and investigation of thermally regulated natural gas storage and transportation systems of a new type using sorbents are considered. The results of full-scale tests of the adsorption compressed gas cylinder system for the GAZ-53 truck are described.

Introduction. Natural gas (methane) is one of the main energy sources and will remain such for the next twenty years. Its enormous reserves are stored in the form of gas hydrates under water in the permafrost zone and exceed the other energy resources of the Earth, including oil, coal, and nuclear fuel. The major portion of natural gas is consumed by the power industry. In terms of its basic properties, methane is on a par with such traditional kinds of fuel as gasoline and diesel fuel and, in some indices, even surpasses them. The advantage of its use is associated with its ecological cleanliness. The use of natural gas as an energy carrier permits considering and solving energy problems in close connection with ecology. There appear favorable possibilities of reducing the formation of solid waste and exhaust gases and eliminating the greenhouse effect. Methane is free of toxic substances that are added, e.g., to gasoline to increase the octane number.

Among the disadvantages of natural gas are its low density and small volume heat of combustion, which leads to the necessity of storing it in large-sized vessels and cylinders. Methane can easily be transported to places of large consumption of it (thermal power stations) through pipelines. It is much more complicated to organize delivery of gas to self-contained small consumers scattered over a large territory, such as dwelling public utilities and small towns and villages. Therefore, the problem of conveying methane by motor, water, and air transport is becoming urgent. Because of its low volume density, gas, when transported in cylinders, has to be compressed to a high pressure (20–30 MPa) or liquified at a low temperature (110 K).

One of the most promising technologies of natural gas storage and transportation is its adsorption on a microporous solid sorbent. In the coming decades, an alternative to standard transport cylinders with compressed gas (20 MPa) will be systems of adsorption storage providing a decrease in the pressure to 2–3.5 MPa without a considerable decrease in the gas volume. In the case where sorbents are used, there is no need for cumbersome and metal-consuming vessels, no energy expenditure for gas compression or liquification, and expenses on compressor equipment decrease.

Works on means of conveying methane in the adsorbed state are being carried out in the USA, Canada, UK, Portugal, Poland, and France. As sorbents for gas storage, activated carbons are favored [1–3]. The theoretically obtained maximum value of the volume density of natural gas storage (the ratio of the gas volume under normal conditions to the cylinder volume) is $195 \text{ nm}^3/\text{m}^3$ at a pressure of 2–3.5 MPa.

Actually, a value of $170 \text{ nm}^3/\text{m}^3$ has been obtained, which is comparable to compressed or liquified gas. The Ford company [4, 5] has developed sorbents and compressed gas facilities which contain, at a pressure of 2 MPa, gas storage equivalent to that of cylinders at a pressure of 15 MPa. The specially designed model of a bound-methane vehicle covers a distance of 100–200 km. In the USA [6], a flat automobile tank filled with carbon sorbent has been made. The volume density of the natural gas storage in it at a pressure of 3.5 MPa is about $150 \text{ nm}^3/\text{m}^3$. An original design of a transportation cylinder for storing adsorbed methane was proposed in [7]. Tests have shown that the appli-

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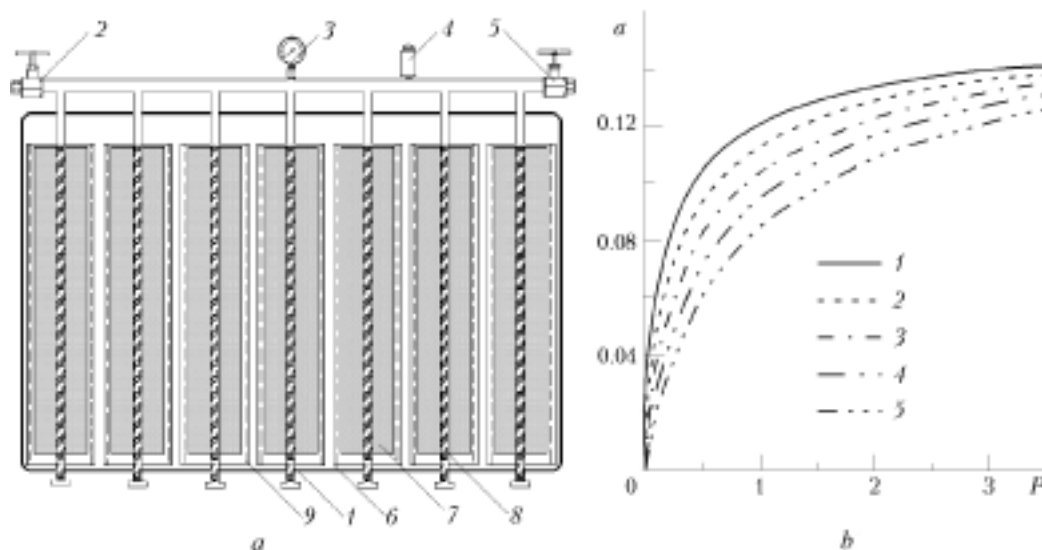


Fig. 1. Sectional cylinder for adsorption storage of natural gas: a) basic diagram; b) isotherms of methane sorption by C107 activated carbon: 1 — $T = 233$ K; 2 — 253; 3 — 273; 4 — 293; 5 — 313. a , kg/kg; P , MPa.

cation of a special aluminum profile gives a number of advantages such as compactness and a small weight compared to common high-pressure cylinders.

Many authors [8, 9] emphasize that the charge and discharge cycles of a cylinder depend on the sorption heat and it is necessary to take into account that any finite rate of adsorption or sorption is associated with the temperature changes in the sorbent volume. In the case of the intensive extraction of gas necessary for functioning of the vehicle, the degree of use of its store can decrease to 50–60%. A counter measure to this is to heat up the sorbent as the gas is extracted via the same system with the aid of which the sorbent was cooled during the filling. In patent [10], the use of alignment in the vehicle cylinder of the air stream channel and the sorbent sections is described.

To increase the degree of extraction of methane from cylindrical vessels, it was proposed to insert an axial perforated pipe for extracting the gas [9]. Two carbon steel cylinders of 23-liter volume were made. As a sorbent, powdery activated carbon with a fairly high methane sorption capacity was used. For example, at an ambient temperature of 293 K and a pressure of 2 MPa its value was $0.134 \text{ m}^3/\text{kg}$. In extracting the gas from the cylinder, the temperature inside the sorbent decreased by 37 K and the desorbed gas volume decreased by 25% compared to the isothermal conditions.

The present paper is devoted to the development of a thermally regulated adsorption natural gas storage system that can provide effective operation of a vehicle in both summer and winter. We have developed, made, and investigated two variants: an experimental sectional cylinder containing heat tubes and a compressed-gas system for the GAZ-53 truck.

Design of the Thermally Regulated Sectional Cylinder and Experimental Facility for Investigating Its Characteristics. The developed 43-liter cylinder of the sectional type is designed for adsorption storage of natural gas at mean pressures of 2–3.5 MPa. The prototype unit (Fig. 1a) was made in the form of a welded construction consisting of seven sections rigidly secured together by two braces and four studs and enclosed in a flat housing 1. The flat form is convenient for arranging and placing the compressed-gas transport facility on a vehicle. On the gas feed and removal header, a charge valve 2, a manometer 3, a safety valve 4, and a valve for connection to the fuel system of the vehicle are located.

As is seen from Fig. 1a, an individual section has a stainless steel housing 6 filled with sorbent 7 in which gas in adsorbed and compressed form is contained. Free gas fills the macropores, whereas adsorbed gas is kept by the molecular interaction forces mainly in the micropores, whose size is comparable to the adsorbed molecules. Due to the high values of pressure in the adsorption space (60–80 MPa), the gas storage density in the cylinder with the sorbent increases. To reduce the influence of the adsorption heat, a heat-exchange element 8 is introduced into each section

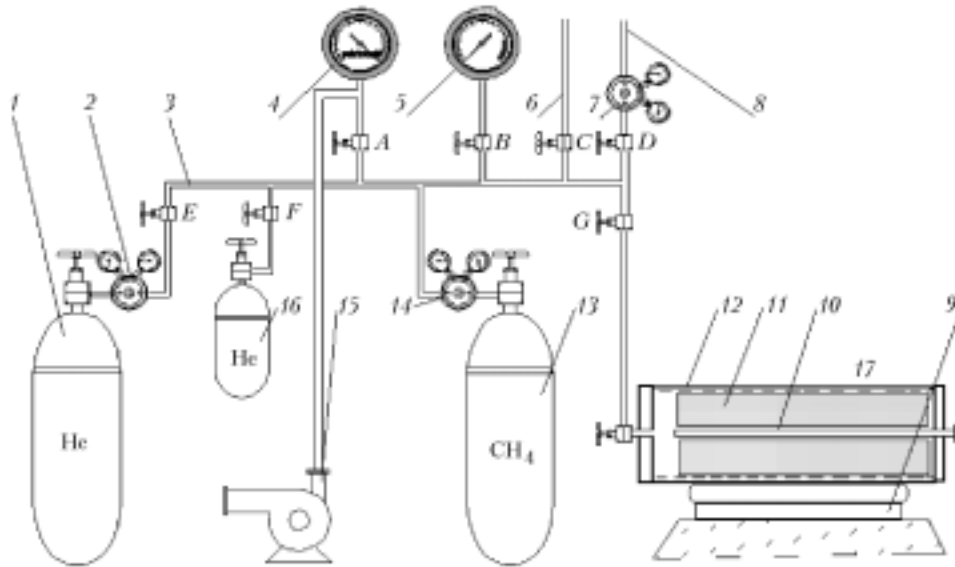


Fig. 2. Basic diagram of the facility for filling and testing the sectional cylinder: 1 — cylinder with helium; 2, 7, 14 — reducers; 3 — dosing unit header; 4 — vacuum gauge; 5 — pressure gauge; 6 — gas release line; 8 — pressure testing line; 9 — scales; 10 — heat pipe; 11 — sorbent; 12 — gas extraction pipe; 13 — cylinder with methane; 15 — vacuum pump; 16 — calibrated vessel; 17 — prototype sectional cylinder for adsorption storage of natural gas; A, B, C, D, E, F, G — valves.

and a perforated pipe 9 for radial conveyance or removal of gas is mounted. In the clearance between them, the sorbent — 207C activated carbon, whose experimental isotherms for methane sorption are given in Fig. 1b, is placed. The header made from a stainless steel pipe 25 mm in diameter ties individual perforated pipes together.

As a heat-exchange element, one may use a heat pipe, a heat siphon, a single-phase heat exchanger, or an electric heater. The heat pipe has advantages over the other devices, since it permits using different kinds of energy (gas flame, electricity, steam, liquid). The air conditioner of the vehicle, and its exhaust gases or radiator also can serve to heat the sorbent. Moreover, the heat pipe — a heat energy "superconductor" a hundred times better than silver or copper — provides homogeneity of the temperature field of the heating surface. The geometric and technical characteristics of the proposed cylinder are given below:

Dimensions, m	1.565 × 0.758 × 0.14
Number of sections in the cylinder, pieces	7
Cylinder volume, m ³	0.043
Outer diameter of section, m	0.076
Inner diameter of section, m	0.073
Sorbent mass in cylinder, kg	20.6
Total volume of charged gas, nm ³	3.9
Operating temperature, K	233–313
Operating pressure in the cylinder, MPa	3.5

The experimental facility for examining the filling and discharge of the cylinder filled with natural gas, whose thermal regime was registered by measuring the heat flows and temperature fields, is shown in Fig. 2. The quantity of adsorbed (desorbed) methane was determined by the gravimetric method. The facility had the following basic elements: an auxiliary unit, a balance, a prototype unit, a dozer, a vacuum system, and a recorder. The dosing part includes: cylinders with helium and natural gas, a header, pressure regulators, and valves. The evacuation system incorporated a vacuum pump and a vacuum gauge connected to the dosing part by a stop valve. The auxiliary part of the facility consisted of a fan with air offtakes and a chamber designed to create around the cylinder under investigation conditions similar to the operating conditions. At the stage of the first filling the prototype unit was placed in a special

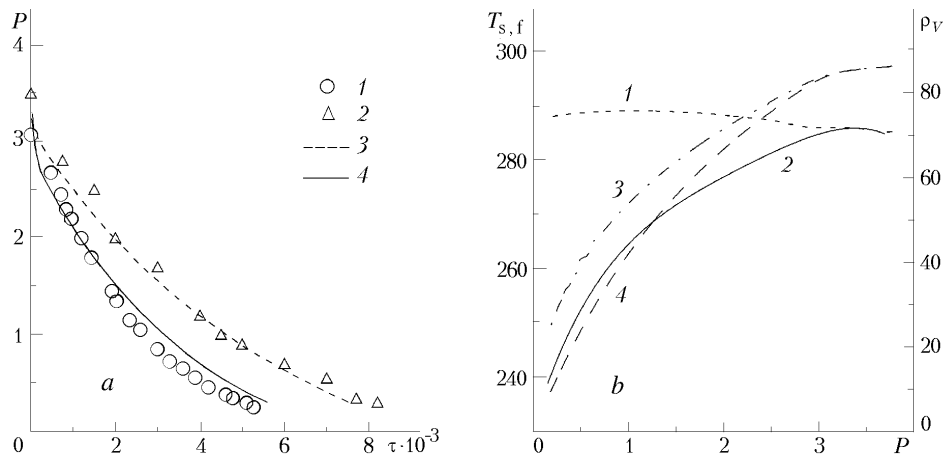


Fig. 3. Changes in the sectional cylinder characteristics in the discharge process at an ambient temperature of 285 K for various values of the additional heat flow (1, 4 — $Q_{h,e} = 280$ W; 2, 3 — 0): a) change in the pressure (lines — calculation, symbols — experiment); b) change in the sorbent mean temperature (1, 2) and in the volume methane storage density (3, 4). $T_{s,f}$, K; ρ_V , nm^3/m^3 ; P , MPa; τ , sec.

housing. Then it was connected to the dosing part of the facility through the valve of connection to the fuel system of the vehicle.

Note that in the course of experiments, to determine the amount of stored gas under given conditions, the sectional cylinder was detached from the facility, weighed, and connected to the facility again. Pressure was controlled by manometer readings. As temperature sensors, copper–constantan thermocouples located inside the sorbent and on the surface of the sectional cylinder were used.

At the preparation stage, the sorbent placed in the cylinder was heated to 420–450 K in order to recover its adsorption properties and remove impurities. Simultaneously with the use of the vacuum system prolonged evacuation of gases from the bulk was performed. The sorbent was considered to be completely freed from impurities if the results of three sequential weighings coincided and after the valve was closed on the vacuum unit line the pressure pickup readings remained constant for 30 min. At the filling stage, the sectional cylinder was connected to the gas main and natural gas was fed portionwise into it through the pressure regulator and the system of manual valves. The process of cylinder filling due to the gas adsorption by the sorbent continued until the pressure in it reached 3.5 MPa and the temperature was stabilized. The investigation of the process of gas extraction was carried out as follows. The prototype unit was connected, by means of the valve for connection to the fuel system of the truck through the gas counter, to the "gas disposal" line. Then the valve was opened and the given flow rate was set by means of the pressure regulator and controlled by rotamer readings. The temperature inside the sorbent could be regulated by the heat pipe. When the pressure decreased to 0.15 MPa, the discharge process was terminated and the cylinder was detached from the facility and weighed to determine the amount of gas remaining.

Analysis of the Experimental and Numerical Results of the Investigation of the Heat-Regulated Sectional Cylinder. Calculated substantiation of the operating conditions for gas cylinders of vehicles is indispensable in creating systems of adsorption storage of natural gas (SASNG). To this end, earlier [11–14], a mathematical model of a SASNG was developed, which represented a system of equations — conservation, energy, kinetics, and equilibrium state equations (isothermal adsorption equations). The kernel of the problem was the two-dimensional nonstationary heat-balance equation with a source term that takes into account the sorption heat and the presence of a heat-exchange element.

In the present paper, partial optimization of the operating conditions has been performed on the basis of the numerical analysis of a 43-liters sectional cylinder. The calculation has been made for one cylindrical section of the SASNG of volume $6.14 \cdot 10^{-3} \text{ m}^3$, holding constant the heat-flow density on the heat exchanger. The summarized performance indices of the cylinder depended on the number of sections and their respective characteristics.

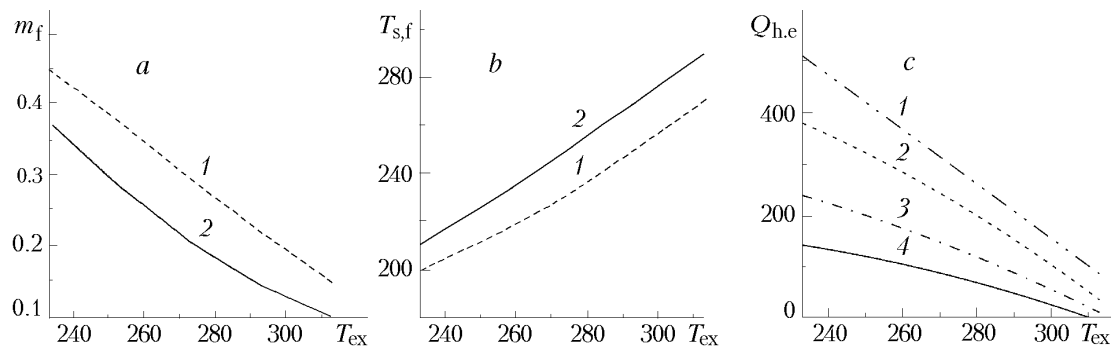


Fig. 4. Sectional cylinder discharge parameters versus ambient temperature: a) dynamic filling coefficient; b) sorbent mean temperature at the end of the process ($P_f = 0.15$ MPa) for various values of the heat exchange coefficient with the environment (1 — $\alpha_{ex} = 0$ W/(m²·K), 2 — 10); c) additional heat flow through heat pipes for various values of the gas flow rate: 1 — $g = 0.4$ g/sec; 2 — 0.3; 3 — 0.2; 4 — 0.1. $Q_{h,e}$, W; T_{ex} , $T_{s,f}$, K.

As a microporous sorbent, 207C activated carbon obtained from coconut shell was used. Its adsorption and thermophysical properties were determined on an *ad hoc* facility [3]. The effective heat conductivity of the carbon at a porosity of 43 and a density of 490 kg/m³ was 0.2 W/(m²·K) and its heat capacity was 1052 J/(kg·K). The empirical coefficients in the equilibrium state and kinetics equations for the 207C carbon–methane pair had the following values: $W_0/b = 0.14$, $B/\beta^2 = 1.977 \cdot 10^{-6}$, $E/R\mu = 890$ K, $K_{s0} = 7.35 \cdot 10^{-2}$ sec⁻¹.

In the calculations, the following basic initial data corresponding to the analogous experimental data were given: the initial temperature was 285 K; the ambient temperature was 285 K; the thermal flow supplied to the sorbent was 0 or 280 W; the number of sections was 7; the mass velocity of methane extraction was 0.433 g/sec; the heat-exchange coefficient at the outer boundary was equal to zero; the initial pressure in the cylinder was 3.5 MPa; the pressure at the end of gas extraction was 0.15 MPa.

Some of the results of the calculations and experiments are presented in Figs. 3 and 4. Figure 3a illustrates the change in the cylinder pressure in the course of the discharge cycle in the absence of heating and with an operating heat pipe. In experiments, the gas discharge at the cylinder outlet was held constant. The process was initiated at a filling pressure P_0 and a dynamic filling factor (the ratio of the initial mass of the gas to its current value) equal to unity. At the end of discharge the cylinder pressure reached a certain threshold P_f given by the pressure regulator and the filling factor showed the portion of the nonextracted gas remaining, $m_f = f(a, P_f, T_{s,f})$. Gas extraction lowers the cylinder pressure, while heating, on the contrary, increases it due to the decrease in the adsorption value and the fact that the sorbent begins again to release the previously absorbed gas molecules. The resulting character of the change in the cylinder pressure depends on these two opposite factors.

Figure 3b shows the curves of the change in the mean temperature of the sorbent and in the volume density of storage in the process of SASNG discharge. It is seen that the sorbent temperature under adiabatic conditions of gas extraction has decreased by 48 K due to the sorption heat absorption. The switching of the heat pipe ($Q_{h,e} = 280$ W) led to an almost isothermal regime of desorption and a decrease in the volume of the remaining gas at a pressure of 0.15 MPa by a factor of about three as compared to the "cool" sectional cylinder. The conveyance of the heat flow to the sorbent produced a noticeable effect on the degree and time of gas extraction from the sectional cylinder.

A certain degree of resistance to the sorbent cooling in the process of desorption can be provided due to the natural heating of the SASNG by the environment. Analysis of the influence on the end parameters of the discharge of the conditions of heat exchange between the cylinder and the environment (Fig. 4a, b) shows that the heat-insulated system with a sorbent ($\alpha_{ex} = 0$, $Q_{h,e} = 0$) at a residual pressure ($P_f = 0.15$ MPa) has up to 20–45% of the initial amount of gas. Enhancement of the heat exchange with the environment ($\alpha_{ex} = 10$ W/(m²·K) made it possible to raise the mean temperature of the sorbent by 10–15 K and reduce the portion of the remaining gas to 10–37%.

In extreme cases (winter time, large gas consumption), to ensure the maximum use of the methane store, it is necessary to choose an optimum regime of heating the adsorption storage system at a given ambient temperature and its fixed consumption. Excess heat supplied to the sorbent will overheat the cylinder and cause a prohibitive increase

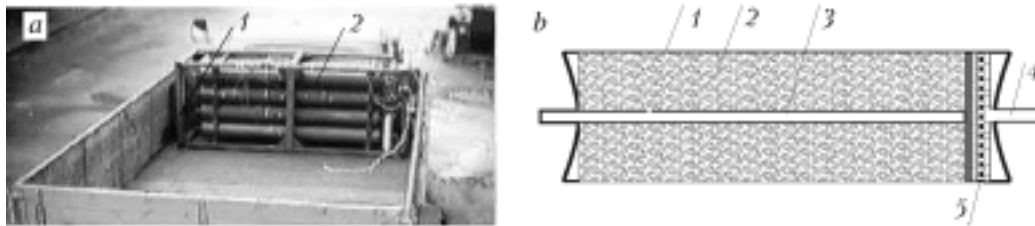


Fig. 5. Adsorption gas cylinder system for natural gas storage installed on the GAZ-53 truck: a) general view; b) diagram of the adsorption cylinder: 1 — housing; 2 — sorbent; 3 — central pipe; 4 — inlet/outlet of gas; 5 — multi-layer filter.

in the pressure, and a low value of the heat flow will not be able to make up for the spontaneous cooling of the sorbent layer as a result of the desorption process. Thus, the aim of the heating is to extend the distance covered by the vehicle by reducing the amount of nonextracted gas remaining.

To determine the optimum operational conditions for the cylinder providing 90% of stored gas consumption, we carried out calculations for various values of the heat flow $Q_{h,e}$ conveyed to the sorbent by the heat pipe, ambient temperatures T_{ex} (equal to the initial sorbent temperatures T_0), and flow rates of the gas. Figure 4c gives the generalization of this series of calculations. At mean values of the gas flow rate $g = 0.3$ g/sec and temperature $T_{ex} = T_0 = 273$ K, an additional heat flow equal to about 300 W is required. An increase in the gas extraction rate and a decrease in the ambient and initial temperatures lead to the necessity of increasing the heat flow from the external source.

Adsorption Gas-Cylinder System for the Automotive Vehicle. The proposed system of natural gas storage in the sorbent-bound state was made, installed, and tested on a GAZ-53 truck (Fig. 5a). The system consisted of twelve 24-liter cylinders with a rigid binding 1 arranged in three assemblies 2. The assemblies are equipped with manual valves for independent disconnection from the fuel system of the truck.

Figure 5b schematically represents the sorption cylinder filled with 207C activated carbon. Located ahead of the cylinder outlet is a three-layer filter consisting of a non-rusting, fine-mesh gauze, a 2-mm-thick layer of woven Busofit-type carbon fiber, an 8-mm-thick layer of carbon felt nonwoven material, and a perforated metal disk. The filter prevented ingress of carbon particles and dust into the fuel system of the truck.

The SASNG was equipped with a safety valve and a standard pressure gauge for pressure control. Thermocouples (from three to twelve depending on experimental conditions) were intended to measure the temperature field in the system charge and discharge process. Their heads were fixed on the housing surface and inside the central pipes of the cylinders. During the experiment, the thermocouples were alternately connected, by means of a commutator, to a portable Shch68009 millivoltmeter and a cold junction placed in a Denar flask.

At the initial stage of preparation, the cylinders were subjected to leakage and strength tests at a pressure of 4.75 MPa and then heated to 337–393 K with simultaneous evacuation for 12 h in order to clear the sorption material from foreign impurities. The SASNG filling with natural gas was carried out in a heated box with a temperature of 283–288 K to the operating pressure of 3.5 MPa in several stages because of the thermal energy release in the adsorption process and the increase in the cylinder temperature. The maximum temperature difference from the ambient temperature was 25 K.

At each stage the filling was terminated when the pressure in the system reached 3.5 MPa. About 80–85% of the total amount of the natural gas stored by the SASNG was filled during 20–30 min at the first stage. The remaining 15–20% was added after the cylinder and ambient temperatures were equalized. Under real service conditions on a vehicle, a single-stage 20-min filling of the gas-cylinder system to a pressure of 4 MPa may be recommended. In so doing, as the cylinders cool down to the ambient temperature, their pressure will decrease to the service pressure. After the system was filled, the truck was brought into an open area for thermostabilization, where in 3–4 h the cylinder temperature became equal to the ambient temperature.

The experimental study of the operation of the adsorption gas-cylinder system of the truck included full-scale tests in the process of truck motion and laboratory modeling of the discharge due to the gas release into the air through the counter. The last experiment was needed to measure the volume of the natural gas stored in the SASNG under given environmental conditions, which can be used as a fuel for the GAZ-53 truck.

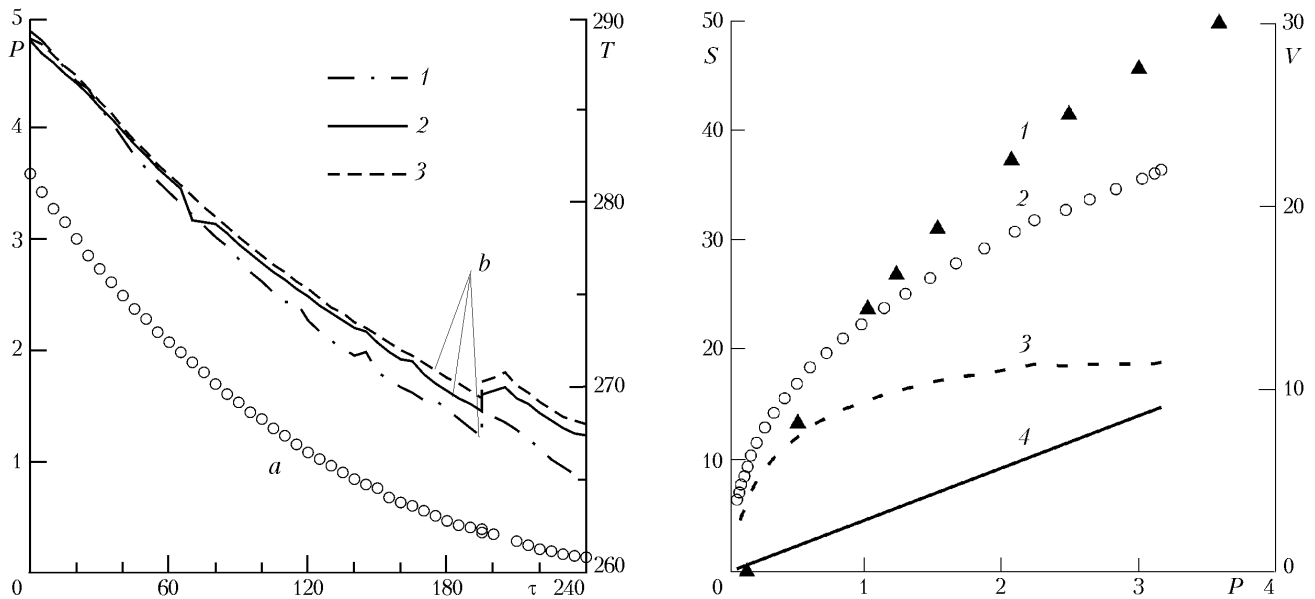


Fig. 6. Change in the pressure (a) and temperature (b) in the adsorption gas cylinder system of natural gas storage installed on the moving GAZ-53 truck: 1 — central cylinder in the assembly; 2 — cylinder located to the right of the central cylinder; 3 — cylinder located to the left of the central cylinder. P , MPa; T , K; τ , min.

Fig. 7. Fuel distance endurance of the GAZ-53 truck (1) and gas volume (2–4) in the gas cylinder storage system versus pressure (2 — in the adsorbed and compressed state, 3 — in the adsorbed state, 4 — in the compressed state in the same volume without sorbent). S , km; V , m^3 ; P , MPa.

The full-scale tests of the SASNG (operating conditions) were carried out with the use of a standard pneumatic circuit for connecting the cylinder gas equipment to the fuel system of compressed-gas-operated trucks. To extract as much gas as possible at low pressures, an additional bypass line was introduced into the circuit around the high-pressure reducer-heater. During the vehicle motion, the temperature on the cylinder surface in each assembly and the pressure in the system were measured at 5-min intervals. The mean ambient temperature was equal to 281 K. Shut-off of the high-pressure reducer, which took 10 min, was carried out 3 h and 15 min after the truck started moving at a residual pressure in the SASNG of 0.39 MPa. As is seen from Fig. 6, the cylinder housing temperatures in practice slightly differed from one another, and this difference was due to their position in the assembly. The temperature of the central cylinder turned out to be 1.5–2 K lower because of the worse conditions of energy exchange with the environment. It should be noted that the 10-min standing idle with a dead engine led to an increase in the pressure and temperature in the gas cylinder system, which continued for some time after the motion was resumed. This is explained by the heating inertia of the cylindrical layer of the sorbent. The residual pressure in the system with a shut-off high-pressure reducer did not exceed 0.13 MPa.

The experiments have shown that the temperature difference inside the cylinder before and after the discharge was equal to 14–23 K and on their surface it was 3 to 8 K and depended on the rate of gas flow from the system. When the fuel was supplied to the truck engine through the high-pressure reducer, 15–20% of the stored gas remained in the system, since the existing standard gas equipment makes its complete use impossible. To provide the maximum extraction of compressed and adsorbed gas after the pressure was decreased to its lowest possible value, the high-pressure reducer was replaced by a pipe 10 mm in diameter. As a result, the volume of gas extracted from the developed transport system, containing 132.5 kg of sorbent, reached 20 m^3 . The distance covered non-stop by the truck at a speed of 12.5 km/h with the gas cylinder system filled once to a pressure of 3.5 MPa was, according to the speedometer, 50 km. Figure 7 gives the obtained dependence of the fuel distance endurance of the GAZ-53 truck on the fill pressure, which is obviously nonlinear and exponential in appearance.

To directly determine the amount of extracted natural gas capable of entering the engine in the discharge process, the gas-cylinder system was disconnected from the fuel system of the truck and connected, through two high- and low-pressure reducers, to a G6-type low-pressure gas counter. The volumetric rate of gas flow through the counter was set close to $4.15 \text{ m}^3/\text{h}$, which corresponds to the conditions of the full-scale tests. The measurement error of the G6 counter in the considered range of flow rates did not exceed 25. Figure 7 reflects the results of the measurements made and illustrates the advantage of the adsorption gas-cylinder system over the standard cylinder with compressed gas in the pressure range under consideration. The pressure–volume curve of the gas in the storage system of the new type is smooth and, unlike the practically linear dependence for the case of compressed gas, is well approximated by the inverse exponential dependence (assuming that the flow rate is constant), which is due to the absorption processes proceeding in it. Obviously, the amount of gas that can be extracted from the gas-cylinder system of the new type at an initial pressure of about 3.5 MPa is almost three times larger than in the case of storage in the compressed state. In the range of low pressures (up to 2 MPa), the advantages become even more pronounced due to the increase in the portion of adsorbed gas.

CONCLUSIONS

The proposed adsorption systems of natural gas storage and transportation are promising for storing large contents of energy (construction of underground and surface natural gas storages for billions of cubic meters) and micro-contents of gas (cylinders of volume from a few liters to hundreds of cubic meters). Such systems can operate at low pressures and temperatures and serve as a "power supply" for self-contained adsorption thermal pumps, high-efficiency boilers, gas stoves, and infrared heaters. The employment in them of a heat-monitoring device on the heat pipes enables one to control the temperature distribution in the sorbent layer, influence the degree and time of gas extraction, and provide optimum operational conditions.

NOTATION

a , current or nonequilibrium value of adsorption, kg/kg; B , constant depending on the size of the micropores; b , constant from the Van der Waals equation; E , activation energy, J/kg; g , mass flow of gas from the cylinder, g/sec; K_{s0} , pre-exponential factor in the approximated kinetics equation; m , dynamic filling coefficient of the cylinder; P , pressure, MPa; $Q_{h.e.}$, heat flow through heat-exchange elements (heat pipes), W; R_{μ} , gas constant, J/(kg·K); T_s , mean temperature of sorbent, K; T_{ex} , ambient temperature, K; T , temperature, K; S , fuel distance endurance, km; V , volume, m^3 ; W_0 , limiting volume of adsorption space expressing the volume of activated carbon micropores; α_{ex} , coefficient of heat exchange with the environment, $\text{W}/(\text{m}^2 \cdot \text{K})$; β , affinity coefficient; ρ_V , volume density of storage, nm^3/m^3 ; τ , time, sec. Subscripts: ex, environment; h.e., heat-exchange element; f and 0, initial and finite values; s, sorbent; μ , molecular weight.

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