ADSORPTION SYSTEMS OF THE NATURAL GAS STORAGE AND TRANSPORTATION AT REDUCED PRESSURES AND TEMPERATURES

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Abstract

Problems of development and research of new type thermally regulated systems for natural gas storage and transportation with solid sorbents utilization are surveyed. Results of natural tests of adsorption system for the automobile GAZ - 53 are described.

KEYWORDS

Natural gas, storage, adsorption, heat pipe, automobile

INTRODUCTION

Natural gas (methane) is one of the primary energy sources and it will keep this position next twenty years at least. Huge reserves of natural gas are stored as gas hydrates under water and in a permafrost zone. The reserves exceed all other energy resources of Earth, including oil, coal and nuclear fuel. The main share of natural gas consumption belongs to the power engineering. The basic properties of methane correspond (and some parameters even surpasses) to such traditional kinds of fuel, as gasoline and diesel fuel. Advantages of the natural gas utilization first of all are connected to an ecological cleanliness. Usage of natural gas as an energy source allows to survey and to work out power problems in close connection with ecological ones. Favorable opportunities for decreasing of a solid waste products formation and harmful automobile emissions, and elimination of a greenhouse effect are framed. The structure of methane fuel does not include toxic substances, which are sometimes added, for example, into gasoline for octane number increasing.

Low density and small volumetric heat of combustion are the disadvantages of natural gas. That results in necessity of its storage in special capacities or containers having the big mass and/or dimensions parameters. Pipelines can easily transport methane to main places of its consumption (thermal power plants). It is much more difficult to organize gas delivery to independent secondary consumers, which are often dissociated on a wide territory, such as housing and communal services, small cities and villages. Therefore the problem of methane shipping with by automobile, water and air transport has an utmost urgency. It is necessary to compress up natural gas to high pressure (20-30 МПа) for its transportation in cylinders because of low volumetric density of gas or to make expensive gas transformation into a liquid state at very low temperature (110 K).

One of the most perspective technologies of natural gas storage and transportation is its adsorption by microporous solid sorbent. In the nearest decades natural gas adsorption systems are going to be alternative to standard transport cylinders with pressurized gas (20 MPa). New systems provide the pressure level decrease up to 2-3.5 MPa without appreciable reducing of the gas storage capacity. In case of the solid sorbents utilization the necessity for bulky, heavy, and metal-consuming vessels is disappeared. Also, expenses of energy on high compression or liquefaction of gas are eliminated and a cost of compressor equipment is cutting down.

Projects on application of methane in an adsorbed state for transportation are intensively conducted in the USA, Canada, England, Portugal, Spain, Poland, France and China [1]. Activated carbons are preferable sorbents for the gas storage [2-4]. Theoretically received maximal value of the storage volumetric efficiency (density) of natural gas (relation of volume of gas under normal conditions to volume of cylinder without sorbent) is 195 nm^3/m^3 at pressure 2.5-3.5 MPa. While another limits are available in literature also: 217 nm^3/m^3 stored volumetric efficiency and 200 nm^3/m^3 delivered value [5]. But to be commercially profitable the adsorption storage is required to have at last 150 nm^3/m^3 efficiency value [6]. This requirement is essential but not only. Economic aspects of adsorption storage vessel development for transportation are discussed in [5].

Value of 150 nm^3/m^3 and more was already achieved in practice for several activated carbon laboratory samples [5] and that is comparable to the high-pressurized gas storages. In "Ford" company [7, 8] sorbents and gas containers were created. The containers are capable to contain the amount of natural gas at pressure 2 MPa same as gas cylinders have at pressure 15 MPa. The specially developed automobile for operation on adsorbed methane has driving range 100-200 km. The flat automobile tank filled with the activated carbon sorbent was made by Atlanta Gas Light Adsorbent Research Group [9]. The storage volumetric density of natural gas for this tank is about 150 nm^3/m^3 at pressure level 3.5 MPa. The original design of a transport cylinder for adsorbed methane storage is described in work [10]. Tests have shown that application of a special aluminum structure gives series of advantages, such as compactness and small weight, in comparison with standard cylindrical high-pressure tanks.

In number of investigations [11–13] influence of sorption heat on gas charge and discharge cycles is emphasized. It is necessary to take into account that any final rate of adsorption (desorption) is close connected to the temperature changes in volume with sorbent. Degree of stored natural gas can decrease up to 50-60 % at high intensive gas outputs. As it was demonstrated in [13] the stored methane capacity is much more sensitive to the level of temperatures and heat transfer then to mass transfer limitations. The authors clime what heat transfer properties are key factor for the success of an adsorbed natural gas system. It is possible to counteract to the negative effects by sorbent heating during gas output with help of a special thermal control system. Same thermal control system can be used for sorbent cooling during natural gas refueling. In the patent [15] overlapping of channels for air flow (temperature control) and sorbent sections natural gas storage is suggested for vehicle tank. Another perspective direction is sorbent bed thermal conductivity increasing by briquetting with the high thermal conductivity inert material (graphite) [13] or without [14].

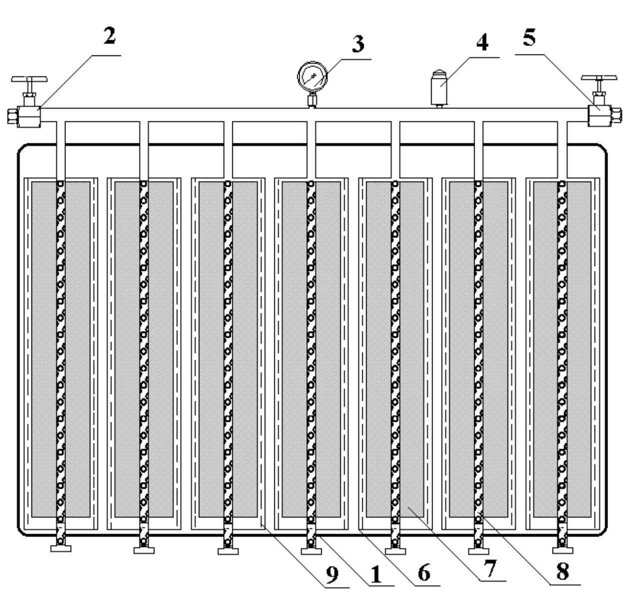
For natural gas degree of extraction maximization it was offered to introduce the axial perforated tube for the gas output from cylindrical storage tanks [12]. It has been made two cylinders from carbonaceous steel with volume 23 liters. The powdery activated carbon with rather high methane sorption capacity was used as the sorbent. For example, capacity was 0.134 m^3/kg at ambient temperature 293 K and pressure 2 MPa. During the gas output from the tank temperature drops up 37 K inside the sorbent and total amount of desorbed natural gas was in 25 % less in comparison with isothermal conditions.

This work is devoted to the thermal controlled adsorption natural gas storage system development. The system is capable to provide an effective work of a vehicle both in summer, and wintertime. Two variants have been designed, made and investigated: the experimental sectional cylinder with the heat pipe, and the gas storage system for Russian lorry GAZ - 53.

THERMALLY REGULATED SECTION TANK DESIGN AND THE EXPERIMENTAL STAND FOR ITS PERFORMANCE INVESTIGATION

The section type tank has volume 43 liters and is intended for the adsorption storage of natural gas at average level of pressures 2-3.5 MPa. The experimental tank (Fig. 1, *a*) was made as an integral welded construction from seven cylindrical sections. The sections are rigidly connected by means of two rods and four dowels and placed in the flat casing *1*. The flat form is rather convenient for configuration and installing of the natural gas transport tank on an automobile. Priming valve *2*, manometer *3*, safety valve *4*, and valve for connection to the automobile fuel system *5* are settled down on the collecting channel for gas input and output.

Every separate section has case from a stainless steel *6*, (Fig.1, *a*) filled by sorbent *7* where natural gas is situated in adsorbed and compressed states. Free gas fills macropores while adsorbed gas is hold by forces of molecular interacting mainly in micropores. Micropore sizes are comparative with adsorbed molecules. Due to high values of pressure in the adsorption space (up to 60-80 the MPa) the storage density of gas in a tank with sorbent increases. For the heat of adsorption effect reduction heat exchange element *8* is inducted in each section and perforated tube *9* providing radial input or output of gas is mounted. Sorbent (activated carbon 207C) is placed in the gap between the heat exchange element and the perforated tube. Experimental isotherms of methane on the activated carbon 207C are shown on Fig. 1, *b*. The collecting channel (stainless steel tube, D_I = 25mm) unites the separate perforated tubes.



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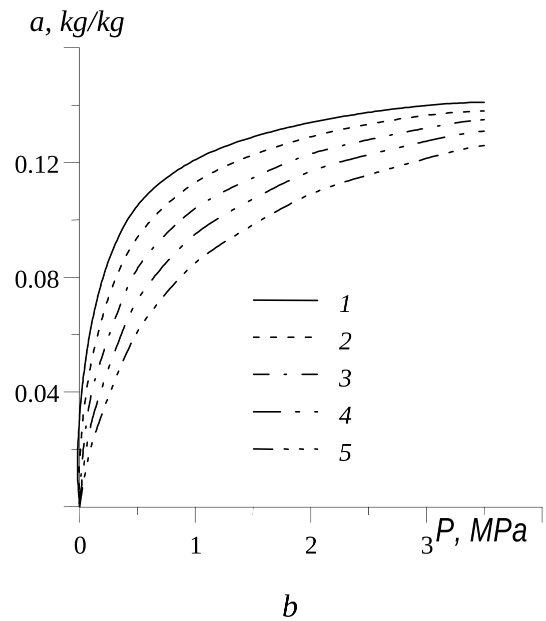


b

Fig. 1. Section tank for adsorption natural gas storage: *a* – principal scheme; *b* – methane sorption isotherms on activated carbon 207C (*1* – *T*=233 K; *2* – 253; *3* – 273; *4* – 293; *5* – 313).

A heat pipe, a thermosyphon, a single-phase heat exchanger or an electric radiator can be used as the heat exchange element. Heat pipes have advantages to other devices. The heat pipe allows us to use different sources of energy (a gas flame, electricity, vapor and/or liquid circuits). An automobile air conditioning system, exhaust gases or an automobile radiator also can serve for sorbent preheating. Moreover the heat pipe (in hundred times better thermal energy "superconductor", than silver or copper) ensures homogeneity of temperature field for heating surface. Geometrical and technical parameters of the designed tank are:

Dimensions, m	1.565 x 0.758 x 0.14
Number of sections in the tank	7
Volume of the tank, m^3	0.043
Outer section diameter, m	0.076
Inner section diameter, m	0.073
Mass of the sorbent in the tank, kg	20.6
Total volume of charged natural gas, nm^3	3.9
Working temperature, K	233–313
Working pressure in the tank, MPa	3.5

The experimental setup for research of filling and discharge of the tank by natural gas is shown on Fig. 2. Investigated tank thermal characteristics were heat fluxes and temperature fields. The amount of the adsorbed (desorbed) methane was determined by a weight method. The experimental setup consisted of the following basic elements: supplementary block, balance, experimental sample, dosing block, vacuum system, and block of a data-acquisition. The dosing block includes in itself two cylinders with helium and natural gas, the collecting channel, pressure reducers, and valves. The system of evacuation was represented by the vacuum gauge and the vacuum pump, which were connected to the dosing block by the valve. The supplementary block of the setup comprised of the fan with airlines and the chamber, intended for creation conditions, which are similar to operating environment around of the tested tank. At the stage of first filling the experimental sample was located in the special casing. Then it incorporated to the dosing part of the setup through the connection to an automobile fuel system valve.

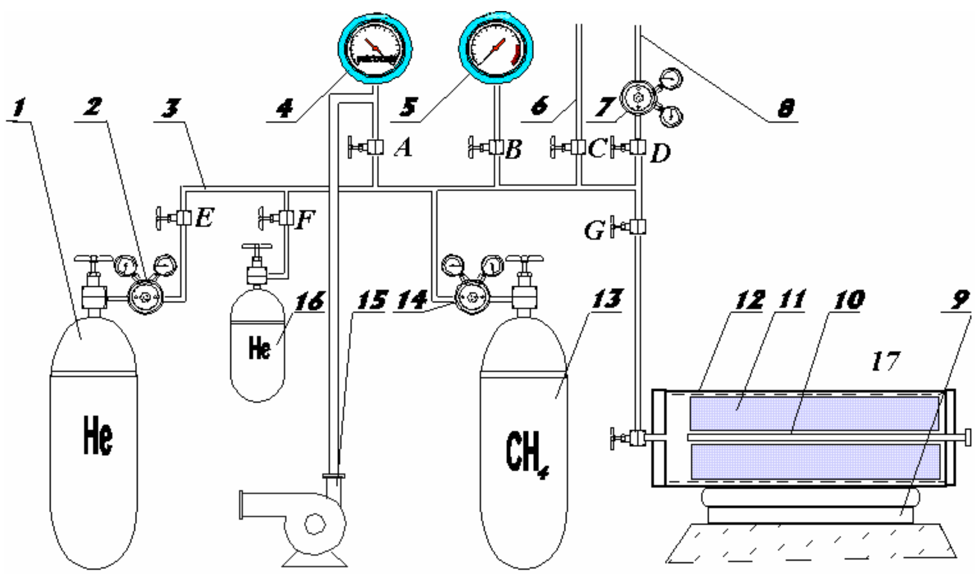


Fig. 2. Principal scheme of the tank charging and testing: *1* – helium cylinder; *2, 7, 14* – reducers; *3* – dosing block collector; *4* – vacuum meter; *5* – manometer; *6* – gas exit line; *8* – pressure test line; *9* – balance; *10* – heat pipe; *11* – sorbent; *12* – gas output tube; *13* – methane cylinder; *15* – vacuum pump; *16* – calibrated volume; *17* – experimental section tank for adsorption natural gas storage; *A, B. C, D, E, F, G* – valves

It should mentioned, that during experiments for definition of adsorbed gas amount at given conditions the section tank was disconnected from the setup and weighed, and then again joined. Monitoring of the pressure was carried out by indications of the manometer. Copper-constantan thermocouples utilized as temperature measuring instruments and were located inside of the sorbent and on a surface of the section tank.

At the stage of preparation the sorbent was located in the tank and the tank was heated up to temperature 430-450 K for adsorption properties restoring and for any admixtures removal. Simultaneously with the heating, the long pump-down of gases from volume was carried out with use of the vacuum system. The sorbent purification was considered finished if results of three sequential weightings were coincided, and if pressure value was unchanged during 30 minutes after closing the valve on the line to the vacuum block. At the stage of filling the section tank was connected to the gas line and natural gas was supplied by certain portions to the tank through the reducer and system manual valves. The tank was totally filled, and the adsorption process was considered finished after pressure set at the level 3.5 MPa and temperature field stabilized. Investigation of the gas output process was conducted by following manner: the experimental sample by means of the connection to fuel car system valve was connected to a line "gas outlet" through the gas counter. Then the valve was opened, and the given gas flow rate was fixed by means of the reducer and inspected under flowmeter indication. A heat pipe could regulate the temperature inside the sorbent bed. When the pressure was lowered till 0.15 MPa the process of discharge was stopped; the tank was disconnected from the experimental installation, and weighed for the measurement of the gas not extracted rest.

THE EXPERIMENTAL AND ANALYTICAL RESULT ANALYSIS OF THE THERMALLY REGULATED SECTION TANK RESEARCH

The analytical justification of the tank operational performances is a necessary part of the investigations in process of natural gas adsorption storage system (NGAS) design and development. With this purpose the NGAS mathematical model [16-19] has been earlier developed. The model based on the system of equations: energy balance, kinetics and equilibrium state (the equation of an isothermal adsorption). The kernel of the problem is the non-stationary heat-balance equation with the source item, which takes into account the heat of sorption and the presence of the heat exchange element.

In the given work the partly optimization of 43-liter section tank operational conditions is executed by means of a numerical analysis. Calculations were carried out for one NGAS cylindrical section with volume 6.14 10^{-3} m^3 for conditions of constant heat flux density on the surface of the heat exchange element. Overall characteristics of the tank operation were yielded in view of tank sections number and corresponding parameters of every section. Activated carbon 207C, obtained from a coconut shell was applied as a microporous sorbent. Its adsorption and thermophysical properties have been defined on the specially developed installation [4]. Effective thermal conductivity of carbon with porosity 0.43 and density 490 kg/m^3 is 0.2 W/mK, heat capacity 1052 J/kgK. Empirical factors for the equations of adsorption equilibrium state and kinetics for working pair active carbon 207C-methane are: $W_0/b = 0.14$, $B/\beta^2 = 1.977 \cdot 10^{-6}$, $E/R_\mu = 890\ K$, $K_{s0} = 7.35 \cdot 10^{-2}\ s^{-1}$.

Following input data which correspond to analogous in experiment have been given in the model: initial temperature - 285 K; environmental temperature - 285 K; input flux was varied from 0 to 280 W; number of sections - 7; mass velocity of the methane discharging - 0.433 g/s; outside heat transfer coefficient was equal to zero; initial pressure in the tank - 3.5 MPa; final pressure - 0.15 MPa. Some results of the analytical and experimental investigations are presented on Figs. 3-4.

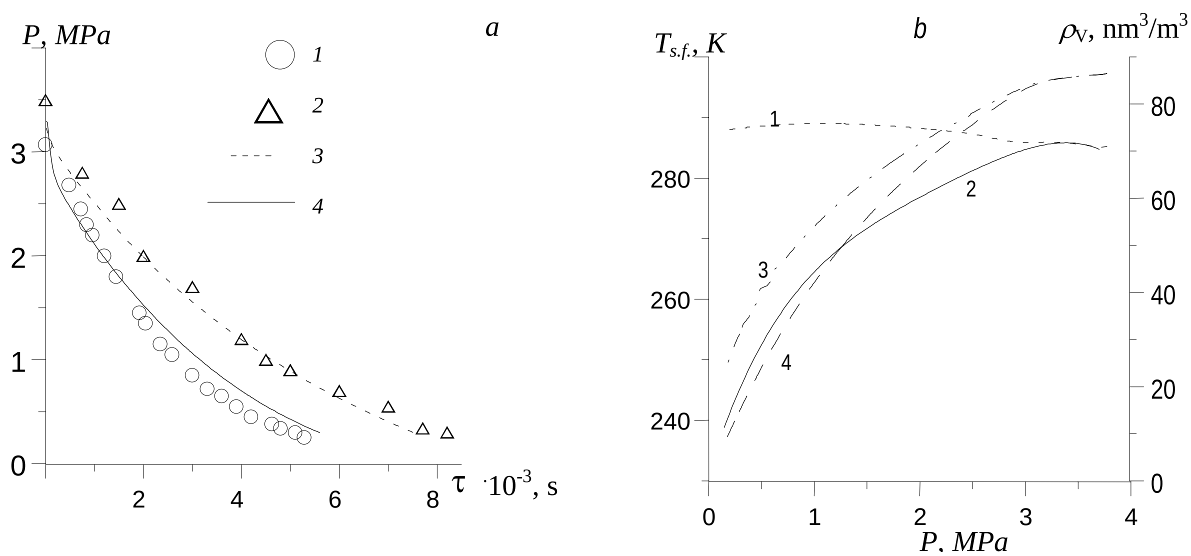
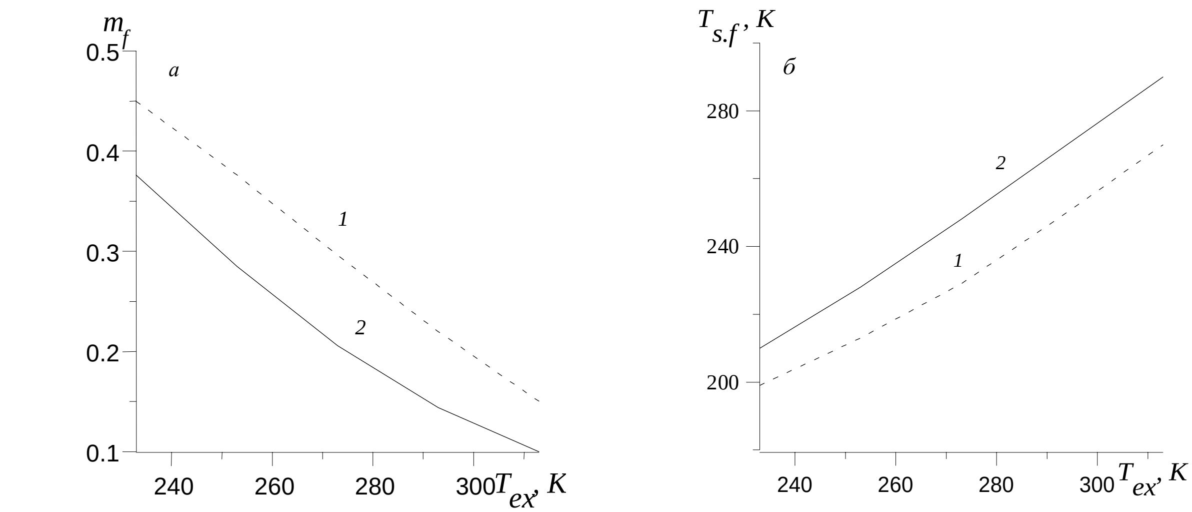


Fig. 3. Section tank operational parameters changing during discharge process at ambient temperature 285 K for different values of additional heat flux (*1*, *4* - $Q_{h.e}$=280 W; *2*, *3* – 0): *a* – pressure change (lines – calculation, points – experiment); *b* – change of sorbent average temperature (*1*, *2*) and volumetric density of methane storage (*3*, *4*).

The gas rate at the tank exit was maintained constant in tests. The process started at initial charging pressure P_0 and at dynamic charging coefficient (the ratio of the initial mass of natural gas to the current gas amount in the tank) equals to one. The tank pressure reached defined threshold P_f, settled by the reducer, at the end of the discharge process. And dynamic charging coefficient was equal a share of not extracted rest of gas $m_f = f(a, P_f, T_{s.f})$. Taking away of gas reduces pressure in the tank, but the heating raises the pressure, on the contrary because of reduction of the adsorption capacity and renewal output of gas molecules, which was absorbed earlier by the sorbent. The Resulting character of the pressure change in the tank depends on these two opposite factors.

Curves of average sorbent temperature change and bulk density of gas storage during the NGAS discharge process are shown on Fig. 3, *b*. It is clear, that sorbent temperature in adiabatic conditions of gas output has decreased by 48 K due to the adsorption heat cooling effect. Turning on of the heat pipe ($Q_{h.e}$=280 W) results to practically isothermal conditions of desorption and to considerable reduction of methane rest in the tank at pressure of 0.15 MPa. The gas rest decreased almost in three times in comparison with "cold" tank case. Heat flux input to the sorbent bed has the noticeable influence on the degree of gas extraction and on the gas discharging time.



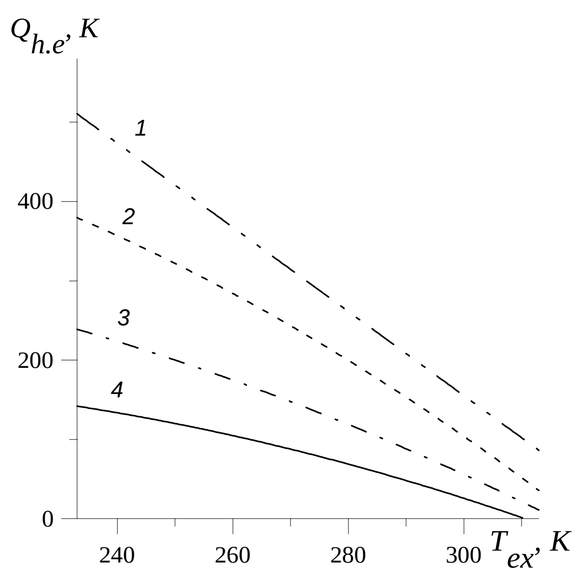


Fig. 4. Section tank discharge parameters dependences on an ambient temperature:

a – dynamic coefficient of filling;

b – sorbent average temperature in the end of the process (P_f = 0.15 MPa) for different values of ambient heat transfer coefficient *1* – $\alpha_{ex} = 0$ W/(m^2 K); *2* – 10;

c – additional heat flux applied by heat pipes for different gas flow rates: *1* – *g*=0.4 g/s; *2* – 0.3; *3* – 0.2; *4* – 0.1.

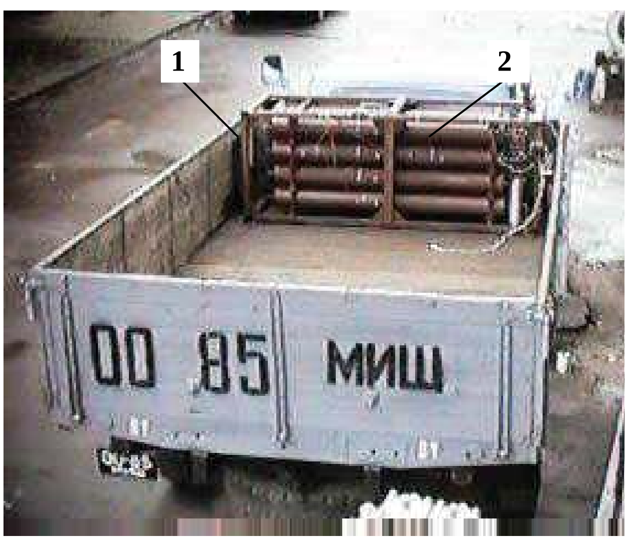
The counteraction to cooling of the sorbent bed during desorption can be provided as a result of NGAS heating by the natural convection from an environment. The analysis of the environment heat exchange conditions influence on resulting discharge parameters of the tank demonstrates, that the heat-insulated system with a sorbent ($\alpha_{ex} = 0$, $Q_{h.e.}$=0) at ultimate pressure P_f=0.15 MPa contains up to 20-45 % of gas initial filling but the heat exchange with an environment ($\alpha_{ex} = 10$ Вт/(м^2·К) has allowed to increase average temperature of the sorbent on 10-15 K and to decrease the share of unused gas up to 10-37 % (Fig. 4 *a, b*).

In special cases (the winter time or/and the increased gas output) it is necessary to select the optimal adsorption system regime of heating at the given environment temperature and at the gas fixed output rate for providing with the stored methane maximum usage. The abundant heat flux to the sorbent bed will result to excessive NGAS heating and the inadmissible pressure increase. But low heat flux cannot compensate a spontaneous cooling of the sorbent due to the desorption process. Thus, heating is required to increase a transport driving range by means of the gas not extracted rest reduction in NGAS.

For the optimal operational NGAS regime (which ensures about 90 % of the stored gas usage) definition, calculations for different applied to the sorbent bed by the heat pipe heat fluxes $Q_{h.e.}$, environment temperatures T_{ex} (there are equal to initial sorbent temperatures T_0) and gas flow rates have been made. Summary of the calculation series is presented on Fig. 4, *c*. At average flow rate value g=0.3 g/s and temperature T_{ex}=T_0=273 K the additional heat flux ~300 W is needed. Growth of the gas output velocity, reducing of an environment temperature and/or the NGAS initial temperature result in necessity to increase the heat flux from an external source of energy.

ADSORPTION GAS STORAGE SYSTEM FOR THE AUTOMOBILE

The suggested system of the natural gas storage on solid sorbents was designed, manufactured, installed, and tested on the lorry GAZ - 53 (Fig. 5, *a*). The system consists of twelve 24-liter cylinders collected in three cartridges 2 with common rigid binding 1. Cartridges are equipped with manual valves for independent switching-off from the fuel system of the automobile.



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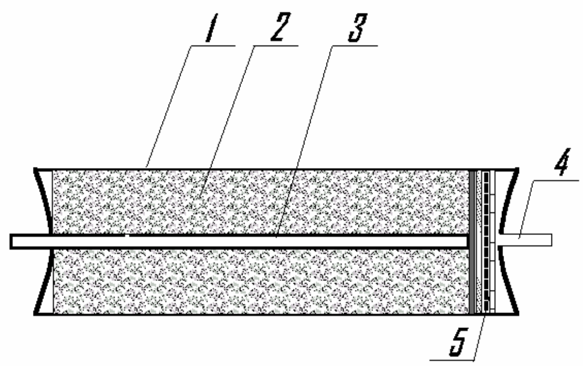

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Fig. 5. Adsorption natural gas storage system installed on the automobile GAZ-53: *a* – general view; *б* – adsorption cylinder scheme (*1* – case, *2* – sorbent, *3* – central pipe, *4* – gas input/output, *5* – multi-layer filter)

The scheme of the adsorption tank filled by the activated carbon 207C is shown on Fig. 5, *b*. Before of the tank output collector the three-layer filter consisting from the stainless steel fine net, carbon fiber "Busofit" (2 mm thickness), carbon felt non-woven material (8 mm thickness) and the perforated metal disk are placed. The filter prevents the penetration of carbon particles and a dust to the fuel system of the automobile**.**

NGAS has been supplied with the safety valve and the high-precision manometer for pressure monitoring. Thermocouples were intended for a temperature field measurement during stages of the filling up and the discharge of the system. The thermocouples were placed on the surface and inside central pipes of the NGAS cylinders.

During the initial preparation of the tank every cylinder was tested for tightness and durability at pressure of 4.75 MPa, and then was heated up to temperature 373-393 K with simultaneous evacuation during 12 hours for the sorption material purification from different admixtures. Filling of NGAS by methane was executed inside a building with ambient temperature 283-288 K up to the working pressure 3.5 MPa by several stages because of the heat energy emission during of the adsorption and the NGAS temperature rise. The maximum temperature decreasing relatively to an environment temperature was 25 K.

The experimental research of the adsorption gas storage system operation for automobile included full-scale tests during of the automobile movement and the laboratory discharge simulation by the gas output to air through the counter. The laboratory experiment was necessary for measurement of the stored natural gas volume, which can be used for the GAZ-53 operation in the given ambient conditions.

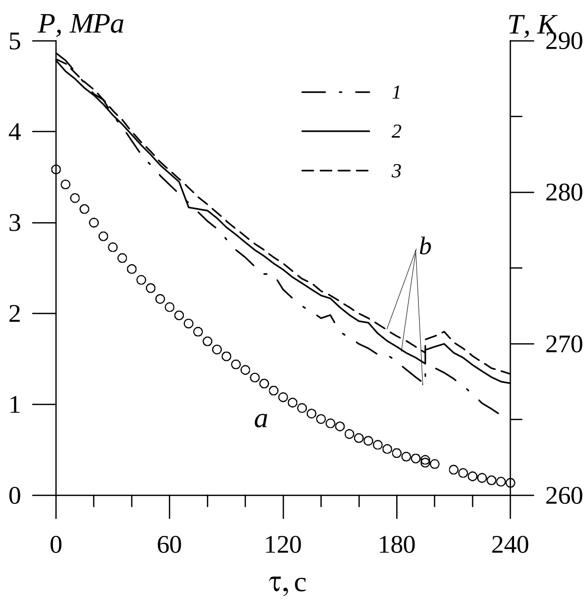

Fig. 6. Change the pressure (a) and temperature (b) in the adsorption natural gas storage system, installed on the running automobile GAZ-53: 1 – central cylinder in the cartridge; 2 – right cylinder from the central one; 3 – left cylinder from the central one.

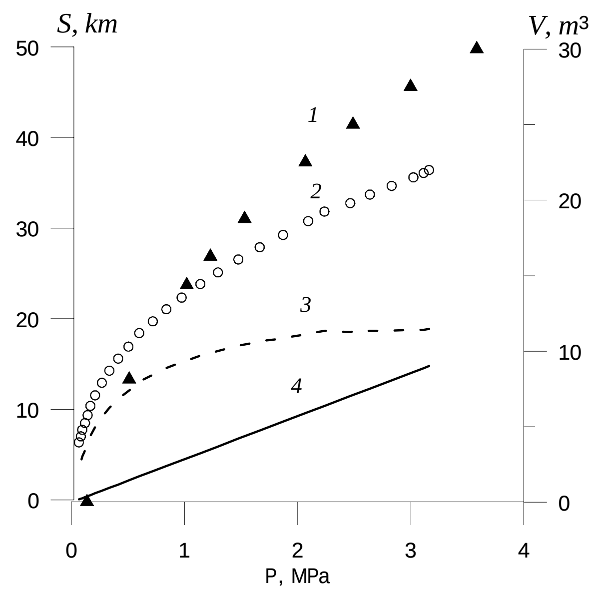

Fig. 7. Dependence of the driving range for the automobile GAZ-53 (1) and of the gas volume (2–4), on the pressure (2 – in the adsorbed and compressed states, 3 – in the adsorbed state only, 4 – in the compressed state for the same volume without the sorbent).

Full-scale NGAS tests were carried out with application of the standard scheme for the natural gas pneumatic connection to the automobile fuel system, which operates on compressed gas. For more complete utilization of natural gas at low pressures the additional gas line around of the high-pressure reducer-preheated was installed in the circuit. During of the car movement temperatures on surfaces of cylinders in every cartridge and the system pressure were measured with 5 minutes time interval. The average environment temperature was 281 K. After 3 hours and 15 minutes since the start of movement at pressure in NGAS 0.39 MPa the high-pressure reducer was switched off. It took 10 minutes. As you see from Fig. 6, temperatures of cylinders practically were not differed from each other a lot, and these variations are caused by cylinder arrangement in cartridges. The temperature of the central cylinder was below on 1.5-2 K because of this one had the worse conditions of a heat exchange with an environment. It is necessary to mark that 10-minute parking with a dead engine caused the increase of the pressure and temperatures in gas storage system, which continued some time after renewal of the car movement. It can be simply explained by the time delay of the cylindrical sorbent bed heating. The final pressure in the system with the switched off high-pressure reducer did not exceed 0.13 MPa.

Experiments have demonstrated, that the temperature difference inside cylinders before and after discharge process was 14-23 K, and on the cylinders surface was 3-8 K and depended on the output gas flow rate. In case if the fuel was fed into the automobile engine through the high-pressure reducer the gas rest in the system was 15-20 % of the initially stored volume because of the existing standard gas equipment cannot ensure gas effective usage. For the maximum extraction of the compressed and adsorbed gas it was necessary to replace high-pressure reducer by 10 mm diameter tube after the pressure decreased up to minimally possible value for the car operation. As a result, the total volume of the extracted natural gas has reached 20 m^3 (system contains 132.5 kg of the sorbent). Continuous run of the automobile with average speed 12.5 km/h was 50 km. On the Fig. 7 obtained dependence of the run storage for automobile GAZ-53 on the system pressure is shown. The curve has the non-linear exponential behavior.

For direct determination of the extracted natural gas amount during NGAS discharge, the gas storage system was disconnected from the automobile fuel system and was joined to gas counter G6 by high and low pressure reducers. The volumetric gas rate through the counter was established 4.15 m^3/hour. It is close to the value corresponding to the full-scale test conditions. The error of the counter G6 in the investigated range of flow rates did not exceed 2 %. The results of the experiments are shown on Fig. 7. It illustrates advantage of the adsorption gas storage systems over standard compressed gas tanks in the given pressure range. The curve of the gas volume dependence on pressure in the new type storage system has the smooth character but the same dependence for compressed gas is linear. The adsorption system line is well approximated by inverse exponential dependence (fixed flow rate). This is explained by the specific adsorption processes, which took place in NGAS. It is obvious, what from the new type storage system it is possible to get the volume of gas almost in three times larger then from a tank with the compressed at pressure 3.5 MPa natural gas. Over the range of small pressures (up to 2 MPa) advantages of the new system become even more appreciable due to the increase of the adsorbed gas share.

CONCLUSIONS

Designed adsorption systems for natural gas storage and transportation are perspective for the power engineering (natural gas underground and ground storage with capacities up to several billions m^3) and for gas domestic and small business industry (tanks in volume from several liters up to hundreds in m^3). Such systems are capable to operate at reduced pressures and temperatures and can serve as "sources of energy" for autonomous adsorption heat humps, high-performance small boiler plants, gas heaters, stoves and infrared heaters. Utilization of heat pipes for the adsorption natural gas storage thermal control allows us to operate a temperature distribution in a sorbent bed, to regulate a degree of gas output and a time of storage discharge, to provide an optimal operational regime.

Nomenclature

a – current or non-equilibrium adsorption capacity, kg/kg; B – constant which depends on micropores size; b – Van der Waals constant; E - activation energy, J/kg, g – mass flow rate from tank, g/s; K_{s0} - exponential multiplier in the kinetic equation; m – dynamic charging coefficient; P – pressure, MPa; $Q_{h.e.}$ – heat flux applied two heat exchange element (heat pipe), W; R_{μ} – gas constant, J/(kgK); T_s – average sorbent temperature, K; T_{ex} – ambient temperature, K; T – temperature, K; S – milage, km; V – volume, m^3; W_0 – volume of adsorption space; α_{ex} – heat transfer coefficient with ambient, W/(m^2K); β – affinity coefficient; ρ_V – volume storage density, nm^3/m^3; τ – time, s. *Indexes:* ex – ambient; h.e. – heat exchange element; I - inner diameter, 0 и f – initial and final values; s – sorbent; μ – molecular weight.

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