Experimental study of heat transfer in adsorbed natural gas storage system filled with microporous monolithic active carbon

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Abstract. The study of the thermal state of the monolithic adsorbent layer and internal heat exchange processes during the circulating charging of an adsorbed natural gas storage system was carried out. The correlation between gas flow mode and the heat transfer coefficient between gas and adsorbent is determined under conditions of mass transfer.

1. Introduction
To reduce the influence of the thermal effects of adsorption and desorption, which reduce the charging and discharging capacity of adsorption gas storage [1], thermal management of the adsorber is often provided [1-7], including circulating charging, in which the gas itself is the heat carrier [6, 7]. In any thermal management systems, heat and mass transfer processes in the adsorber play a key role in ensuring efficient charging. Often in the scientific literature there are diagrams and dynamic temperature dependences at different location points of the adsorber [3, 6, 8, 9], but rarely such data are analyzed to obtain practical results. Due to its technical complexity, circulating charging is the least studied among other methods of thermal management.

2. Materials and methods

2.1. Monolithic microporous carbon adsorbent
In this work industrial granulated activated carbon AU-1 is used. In [10] it was shown, that AU-1 could be potentially used in ANG systems due to high activity to methane, mechanical strength and the possibility to increase packing density by shaping under pressure [11, 12], which ensures the maximum indicators of the volumetric storage capacity of the system.

The technology of manufacturing samples (14 pcs.) of shaped cylindrical monoliths from AU-1 activated carbon is described in [13]. Compared to the original granulated adsorbent, the packing density of the shaped material was increased from 380 to 705 kg/m³. The average diameter and thickness of the monoliths were 196 and 101 mm respectively. To reduce the hydraulic losses of the samples 37 perforating channels with a diameter equal to 4.5 mm were made in the monoliths.
Figure 1 shows a photograph of samples of a monolith adsorbent before the installation in adsorption tank of 51 liters inner volume. Figure 2 shows the geometry of the arrangement of channels in the monolith. Estimated occupation of inner space of the tank by the monoliths is about 80%. The total surface area of the adsorbent monoliths: outer cylindrical surface is $0.86 \text{ m}^2$, the edge surface is $0.82 \text{ m}^2$, the surface of the channels is $0.72 \text{ m}^2$. Thus, such a number of holes play the role of a flow equalizer rather than increasing the heat transfer surface area.

2.2. Adsorptive
Natural gas with the following composition was used in the experiments, vol.: 96.1% methane, 2.2% ethane, 0.8% propane, 0.6% nitrogen and other impurities less than 1%. Theoretical calculations and experimental data analysis were carried out on the basis of 100% (pure) methane.

2.3. Experimental Setup
To study natural gas charge and discharge processes a special multifunctional experimental setup was developed and manufactured. The scheme of this experimental setup is presented in [14].

To control the temperature distribution inside the heat-insulated adsorber, temperature sensor tubes are installed in it. The installation diagram of these sensors is shown in Figure 3. Sensitive elements of temperature sensors 1, 6, 8, 9 are located along the axis of the adsorber, the sensitive element of the sensor 7 is located at the entrance to the adsorber. Temperature sensors 2-5 are located to evaluate the radial temperature gradient.

The circulating circuit of the experimental setup includes a supercharger and heat exchangers for removing heat into the environment (stage 1) and to the coolant (stage 2) interacting with the cooling unit. The splitting of the gas flow in adsorber tank was carried out in order to reduce the hydraulic resistance of the adsorbent bed.
2.4. Methods
Determination of the heat transfer coefficient in this experiment was not a target task and, in fact, is a by-product of the study of integral processes of charging a full-size adsorber. The value of the heat transfer coefficient is calculated by indirect methods: the heat flux removed from the adsorbent is determined from the adsorbent temperature change using thermodynamic approaches described in detail in [13, 15]; the logarithmic mean temperature difference is determined from the experimentally determined temperatures of the adsorbent layer and the inlet gas flow with outlet gas temperature calculated from the heat balance and with corrections using the adsorbent temperature gradient along the adsorber axis and in the radial direction. In order to exclude heat transfer in the gap between the monoliths and the walls of the adsorber, in which, in addition to the gas and the adsorbent, the metal parts of the adsorber and the environment participate, only the adsorber regions close to the axis are considered.

The Reynolds number of the flow can be reduced both to the total section of the adsorber $Re_f$, and to the "narrow" section, taking into account the space occupied by the adsorbent $Re_c \approx 0.48 \cdot Re_f$ (the hydraulic diameter is equal to the channel diameter 4.5 mm). Due to the complex nature of heat transfer, the correct choice of the Reynolds number is difficult: $Re_f$ is more reasonable for heat transfer with the edge surface, and $Re_c$ corresponds heat transfer in the channels.

3. Results and discussions
Figure 4 shows the dependence of the temperature at various points of the adsorber on time in the case of charging at a pressure of 3.5 MPa and a gas flow rate of 12 m$^3$/h. There is a clear heterogeneity in the cooling of the one half adsorber compared to the other: the adsorbent in the area of sensors 1 and 6 is cooled much faster than in the area of sensors 8 and 9. Ensuring the uniformity of flow distribution seems to be one of the most difficult technical problems in real systems: dusting the adsorbent, clogging filters, etc. can significantly distort the distribution of flows. It was found that the approximate ratio of gas flow rates is 2:1, which on the one hand is not so much different from the expected equal ratio; on the other hand, it completely changed the nature of the adsorber cooling and, probably, increased the duration of charging.

To study heat transfer processes, a series of 13 experiments with charging was carried out: 8 experiments with a constant gas flow rate and 5 more experiments with a constant flow mode (Reynolds number at the inlet to the adsorber). Figure 5 shows the distribution of the calculated heat transfer coefficients from the gas flow mode ($Re_f$) in a logarithmic coordinate system. Specific points in the figure represent the calculated instantaneous values, separated from each other by time intervals of at least 120 seconds. Despite the wide spread in the values of the calculated heat transfer coefficient, the figure shows an obvious pattern between it and the Reynolds number. The results do not show the points corresponding to the calculated logarithmic mean temperature difference between the adsorbent and the gas less than 5 K and more than 30 K, i.e. areas with the least reliable results are excluded. Also, about 5% of the roughest points were eliminated, i.e. the results are presented with a confidence interval of 95%. The values of the heat transfer coefficients in logarithmic coordinates are well approximated by a straight line. However, it is likely that the dependence is more complicated, since lower Reynolds numbers correspond to the laminar–turbulent transition. The tangent of the tilt angle of the straight line in the graph coordinates is approximately in the range from 1.4 to 1.7 (the line in the figure has a value of 1.55). The high value of the tangent of the angle is probably due to the effect of mass transfer on heat transfer: with intense heat transfer, both the cooling rate of the adsorbent and the mass transfer between the gas and the adsorbent increase, which in turn accelerates heat transfer. The distribution shown in Figure 5 should be used in practice with caution, since it correspond to the specific geometry of the adsorber and the structure of the adsorbent.
Figure 4. Dependence of the temperature of the sensors (1, 6-9) on time during circulating charging. Charging mode: pressure 3.5 MPa, gas flow rate 12 m$^3$/h.

Figure 5. Distribution of the calculated values of the heat transfer coefficients from the flow mode $Re_f$. Dots are experimental values, line is an approximation.

4. Conclusions
The correlation between the heat transfer coefficient and the gas flow mode for circulating isothermal and low-temperature charging has been determined. This dependence can be used for practical design and simulation of circulating charging ANG systems, for example, to determine the charging duration, the required inlet gas temperature, etc.

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