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Adsorption system with heat pipe thermal control for mobile storage of gaseous fuel

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ABSTRACT

Mobile storage of gaseous fuel is the design bottleneck in a commercialization of vehicles powered by natural gas and hydrogen. This paper is devoted to the questions of development of the system of effective accumulation and safe storage of natural gas in a coupled state by activated carbon fiber based on commercially available "Busofit TM-055". Original experimental setup was developed for carrying out investigation of the processes of adsorption of gaseous fuel in the different carbon adsorbents and for simulation of gas charging/discharging of a storage vessel. The experimental data for methane equilibrium adsorption in a wide temperature range between 233 and 313 K and pressure up 5 MPa are obtained and analyzed from the standpoint of the theory of volume filling of micropores and the linearity of the adsorption isosteres. In the supercritical temperature region modified Dubinin–Astakhov equation are proposed for the prediction of the isotherms and isosteric heat of adsorption.

In a second part, the basic settings of CFD model in terms of adsorption equilibrium, kinetics and coupled heat and mass transfer are described and results are discussed in regard to measures. The heat pipes are considered as a basic element of the thermal control for the storage vessels. A more detailed study of dynamic behavior of thermally controlled storage has been performed for constant mass flow rate and constant pressure of the methane at the inlet of the adsorber-vessel.

1. Introduction

Natural gas and hydrogen have the potential to be a very attractive alternative energy carrier. Usage of natural gas as an energy carrier allows to survey and to work out power problems in close connection with ecological ones. Mobile storage is a major barrier to the widespread use of non-petroleum fuels (natural gas, biogas and hydrogen) in power systems and transportation. Because of the gaseous nature of these fuels, it must be stored onboard a vehicle in either a compressed gaseous or liquefied state. Natural gas (NG) is made up of about 95% methane – a gas that cannot be liquefied at ambient temperature. It is liquefied under pressure of 1–2 MPa at 111.65 K. Liquefied NG requires the use of complex and expensive liquefaction equipment, thermos-like tanks and significant energy consumption for the liquefaction and degasification. Compressed NG is currently the prevailing storage technology. Compressed NG is stored at a high pressure, usually 20–25 MPa and demands very robust specially-designed cylindrical tanks, which use a lot of space and are heavy. In addition, gas compression requires expensive multi-stage high-pressure compression technology [1,2].

Adsorbed NG storage technology is a viable alternative to compressed gas having higher energy density, less extreme operating conditions and less danger of explosions. It has quite a few promising advantages over existing methods. New systems provide the pressure level decrease up to 2–6 MPa without appreciable reducing of the gas storage capacity. Projects on application of natural gas in an adsorbed state for transportation are intensively conducted in the USA (AGLARG), Japan (Honda Motors), Italy (EU FP5 LEVINGS), Brazil (Brazilian Gas Technology Center), China (China University of Petroleum) and et al. in the nineties of the previous century [1–5].

Traditional carbon materials because of their low cost and weight are considered for a long time as the suitable environment for reversible storage of hydrogenous gas due to physical adsorption. The principles of NG adsorption have been around for decades however no one had succeeded to develop commercially viable technology until now. In 2000 the U.S. Department of Energy (DOE) set a target for methane sorption systems to achieve 180 v/v or

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117 g/L at 3.5 MPa and 298 K [2]. In spite of the DOE has not generated new goal for the storage of methane since 2001 adsorbed NG storage technology remains an area of interest to the small businesses and consumers. The phenomenon of physical adsorption is essentially accumulation of the undissociated gas molecules on a surface of microporous carbon fibers or particles. This property is due to the fact that the carbon could be prepared in a very fine powdered or fiber form with highly developed porous structure and due to specific nature of the interactions between carbon atoms and gas molecules. The total amount of the adsorbed hydrogen or methane strongly depends on the pore geometry and pore size distribution as well as the storage pressure and temperature levels [6,7]. In the early 21st century development of hydrogen power engineering had promoted to the revival of interest in the systems of the adsorption storage of natural gas. It was found that the same class of materials (activated carbon) has great potential, both for the storage of hydrogen and methane. Two distinctive characteristics of activated carbons are the large amount of micropores and the resulting large surface areas (1500–3500 m^2/g). Both of these attributes make activated carbon attractive as a gas storage media [8].

Perspective materials used as fillers in storage systems must conform to the requirements of reversibility and durability, moderate adsorption enthalpy, fast gas kinetics and affordability. To reach a high adsorption capacity on a volumetric basis condition, carbon adsorbents must have the optimized and balanced characteristics. Some of these characteristics include [6–8]: 1) high microporosity, since micropores are responsible for most of the adsorption of small molecules like methane; 2) high compactness, which contributes to increase the bulk density and, consequently, the volumetric storage capacity; 3) narrow pore size distribution centered around 0.8–1.1 nm, which optimizes the density of the adsorbed phase; 4) relatively low mesoporosity, since mesopores reduce the bulk density but they facilitate the kinetics of the adsorption processes. Advanced systems are being considered for hydrogenous gas storage: adsorption on different nano- and microporous carbon materials or metal and complex hydrides, metal organic frameworks. Therefore the sorption technology of storing is closest to the practical realization for both methane and hydrogen.

The storage systems, based on an adsorption process however, are submitted to intrinsic thermal effects that can significantly influence the system behavior [9,10]. The thermal effects of the process of adsorption are closely related to the external heat transfer of the storage system, thermophysical properties of structural materials, and to the organization of heat transfer in the adsorbent bed. As follows from the investigations performed in Refs. [11–14], the temperature of the storing system is an important factor that determines the kinetics and thermodynamics of the processes of filling and extraction of gas. This causes the necessity of regulating the supplied and removed heat fluxes with the help of special thermal controlling systems. The key for the success in finding an efficient storage system is the selection of a suitable adsorbent and thermal management.

It should be mentioned that heat or cold must be delivered as uniformly as possible to all the adsorbent portions placed in the reservoir. The currently employed methods of heating with the use of electrical heaters, water coils, hot exhaust gas tubes, etc. do not provide the required heat uniformity in the adsorbent bed and have a very low efficiency. The situation becomes more complicated because only direct blowing of the vessel or using expensive and complex water heat exchangers or special refrigerators can ensure the delivery of cold. The heat pipes proposed as a basic element of the thermal control for the storage vessels are known in principle. There are several Japanese applications (05-092369, 60042867) and patents (JP59003001, JP622204099), as well as a US patent (4,599867) and Belarus patent (Pat. BY, INt.Cl. FO2M 21/4, № a 19991158), concerning the employment of heat pipes for hydrogenous gas storage systems. Heat pipes are highly reliable and efficient heat transfer devices considered for many terrestrial and space applications. This device uses the latent heat of vaporization (condensation and evaporation) of a working fluid to transfer relatively large amounts of energy over a long distance with a small temperature drop. Heat pipes can easily be implemented inside adsorption storage vessels due to their flexibility, simple manufacturing technology [11,15].

The paper is devoted to the problem of development of the safe storage of gaseous fuel in coupled state. Original experimental setup and design of thermally regulated adsorber-vessel are presented. A theoretical and experimental study is conducted to determine the methane adsorption by activated carbon fiber (based on commercially available "Busofit TM-055) in the region of supercritical temperature and moderate pressure (3–5 MPa). Natural gas is modeled as pure methane. Another interconnected work objective is development of tools (based on CFD modeling) able to predict the thermal effects that occur in the system with adsorbent and heat pipe, and to propose solutions for natural gas storage in manifold conditions. Successful implementation of the objectives of the study is of interest for large manufacturers, small businesses and consumers by safe and environmentally friendly transport and energy applications.

2. Experimental setup and test vessel

The schematic diagram of the experimental setup is shown in Fig. 1, *a*. It is intended for carrying out experimental investigations of steady-state and dynamic regimes of the processes of adsorption of the natural gas (methane) on the carbonaceous materials.

The setup consists of the following functional blocks [16]:

– a block for holding high-pressure cylinders (~20 MPa) with tested gases. It is a metal cabinet for standard high-pressure cylinders with tested gases and a cylinder with helium;
– a calibration block of calibrating cylinder with an air heat exchanger for thermostatting, a system of valves for manual and automatic filling of cylinder, temperature and pressure sensors; the calibrating unit ensuring preliminary storage of a specified amount of tested gas;
– a block of flow meters which consists of a system of cocks and valves, two Flow-Lo and Flow-Hi devices for different measurement ranges and for controlling the volumetric and mass flow rates of the adsorbed and desorbed gas;
– a vacuum pumping block consisting of an oil-free spiral and turbomolecular pumps with cocks and an exhaust line (into the ventilation); it is intended for removing air and other gases from the setup;
– a block of adsorption that includes a test vessel with a cut-off cock, a pressure sensor P_s, thermocouples, joints, and a HL85 thermostat. The vessel filled with a adsorbent is equipped with a double-pipe heat exchanger, and a thermostatic system; a separate detachable electric heater is mounted to perform desorption on its outside wall;
– a system of acquisition and analysis of experimental data with an Agilent HP34980A multimeter, communication cables, and a computer.

The combination of the positions of the switches of cocks ensures the pumping out of the setup loop, of the adsorbent-filled vessel, performing adsorption and desorption that correspond to gas charging into the adsorbent-filled vessel and its discharging.