Methane/natural gas storage and delivered capacity for activated carbons in dry and wet conditions

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Abstract

Methane/natural gas storage and delivered capacity for three different activated carbons in dry and wet conditions were measured. In all tests the temperature of the bed was maintained constant at 277.15 K and pressure was increased up to 10 MPa. Natural gas storage capacity was less than methane storage capacity in dry conditions for all the three activated carbons tested, while the gas delivery was almost the same. One of activated carbon tested (NC120) showed the possibility of hydrate forming for pressures higher than 4 MPa but the amount of gas stored still was less than the amount stored in dry conditions over the whole range of pressure. The analysis of the gas delivered at each pressure steps shows that considerable amount of heavy components do not come out from the bed even at very low pressures in both dry and wet condition tests. Repeatability of the sorption/desorption processes – vital for possible commercial/industrial use – has been examined over various cycles.
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1. Introduction

Gas storage in activated carbon (ANG) has been widely investigated as an alternative to Compressed Natural Gas (CNG) and Liquefied Natural Gas (LNG) technologies for storage and transportation of natural gases [1–3]. LNG technology needs very low temperatures (around 108.15 K). As such, standard low temperature storage methods are not suited to the everyday use of natural gas as a fuel, particularly for non-stationary uses. The main disadvantage of using CNG technology for transportation purposes is the need for expensive and heavy high-pressure storage vessels designed in a specific shape. Replacement of high-pressure compression with ANG reduces the pressure dramatically. This reduction in pressure allows using lighter, cheaper and safer onboard storage reservoirs and vehicle can refuel directly from a high-pressure natural gas pipeline.

The results reported from research in this area show that the target value of 150 STP volumes of gas per volume of storage vessel (V/V) deliverable at 3.5 MPa and room temperature [4], is still hardly reached. Recently some investigators have tried to increase the amount of methane stored in activated carbon by coupling adsorption with gas hydrate formation through pre-adsorbed water on the porous material [5–8]. In most of the past ANG studies (in wet or dry conditions), methane is used as the test gas to measure the amount of gas stored. However, in actual systems, natural gas will be used as fuel for storage and data for natural gas storage and delivery on activated carbon are scarce. The high molecular weight species present in natural gas are more strongly adsorbed than methane, especially in the low-pressure region [9]. These high molecular weight components will be preferentially adsorbed on activated carbon and therefore the amount of gas that can be delivered by the storage system will decrease. In wet

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conditions, the presence of higher molecular weight species affects the adsorption process and also the temperature and pressures required to form hydrates.

In this work, natural gas sorption/desorption processes on three different types of commercial activated carbons have been investigated at 277.15 K and up to 10 MPa in dry and wet conditions. The delivered gas at each stage has been analyzed by gas chromatography. For comparison purposes the same work has been also carried out using methane. The repeatability of the above over a number of formation/regeneration cycles has been tested and the composition of the delivered gas has been assessed by online gas chromatographic analysis.

2. Experimental

2.1. Experimental devices

2.1.1. Sorption/desorption apparatus

A cylindrical cell (schematic diagram shown in Fig. 1a) made of stainless steel, is used to measure the amount of gas uptake by activated carbon. The cell volume is about 280 cm^3 and it can be operated up to 40 MPa between 243.15 K and 323.15 K. The equilibrium cell is held in a metallic jacket heated or cooled by a constant temperature liquid bath. Circulating coolant from a cryostat within the
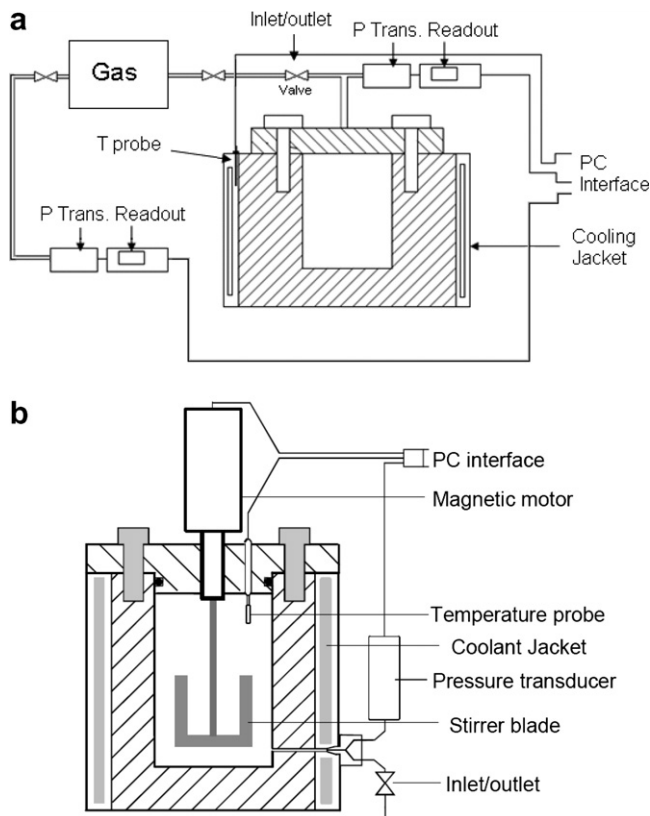

Fig. 1. Experimental setups: schematic diagram of the activated carbon equilibrium cell (a) and of the gas hydrate condition measurement apparatus (b).

jacket surrounding the cell controls the temperature. The cryostat is capable of maintaining the cell temperature to within 0.1 K. To achieve good temperature stability, the jacket and the connecting pipes are insulated. A platinum resistance probe measures the temperature and is connected directly to a computer for direct acquisition. The pressure is measured by means of a strain gauge pressure transducer mounted directly on the cell and connected to the same data acquisition unit. This system allows real time readings and storage of temperatures and pressures throughout the different isothermal runs. Pressure measurement uncertainties are estimated to be within ±0.7 kPa in the operating range. For gas delivery measurement experiments, a gas-meter is fitted to the bottom valve. The gas-meter is equipped with pressure, temperature and volume measurement devices.

2.1.2. Analytical work

The analytical work was carried out using a GC (VARIAN model CP-3800) equipped with a thermal conductivity detector (TCD) and a flame ionization detector (FID) connected to a data acquisition. The analytical column is Hayesep T 100/120 Mesh column (silcosteel tube, length: 1.5 m, diameter: 1/8 in.). The FID was utilized to detect the hydrocarbons and the TCD to detect nitrogen and carbon dioxide. They were repeatedly calibrated by introducing known amounts of standard gas mixture through a gas syringe in the injector of the GC.

2.1.3. Hydrate dissociation apparatus

Another equilibrium cell was used to measure the hydrate dissociation conditions for the gas mixture used in this work. The cylindrical cell (schematic diagram shown in Fig. 1b) is made of stainless steel. The cell volume is about 2500 cm^3 and it can be operated up to 40 MPa between 243.15 K and 323.15 K. A constant temperature liquid bath controls the cell temperature within 0.1 K by circulating the coolant through a metallic jacket. A platinum resistance probe monitors the temperature inside the cell and the pressure is measured by means of a strain gauge pressure transducer mounted directly on the cell. Both the temperature and pressure measurement equipments are connected to the same data acquisition unit. To achieve a faster thermodynamic equilibrium and to provide a good mixing of the fluids, the cell is equipped with a magnetic stirrer with adjustable rotation speed.

2.2. Experimental procedures

For the dry tests, the activated carbon samples were kept in the oven for 12 h at 423.15 K. A known amount of activated carbon is then loaded directly into the cell and vacuum is applied to the cell for one hour. A 300-cm^3 gas cylinder is first connected to the vacuum pump and then pressurized with the desired gas (methane or natural gas). This gas cylinder is then directly connected to the loading valve of the equilibrium cell. The pressure inside