



Research Paper

Methane adsorption on the graphene sheets, activated carbon and carbon black



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HIGHLIGHTS

- Langmuir-Freundlich model is suitable for methane adsorption equilibrium.
- The GS and activated carbon have similar isosteric heat of methane adsorption.
- The carbon black has the smallest isosteric heat of methane adsorption.
- The state of adsorbed methane molecules on the carbon black is the densest.
- The GS and activated carbon have similar interaction energy among adsorbate.

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ABSTRACT

Behavior of methane adsorption on typical carbon based materials was comparatively studied for development of storage medium for adsorbed natural gas (ANG). Three kinds of carbon materials, graphene sheets (GS), activated carbon and carbon black, which respectively has a specific surface area about 300 m²/g, 1118 m²/g and 76 m²/g, were selected for adsorption equilibrium testes within temperature-pressure range from 253.15 K to 293.15 K and 0 to 8 MPa. Langmuir-Freundlich (L-F) equation and a lattice theory based adsorption model were employed to determine the isosteric heat of adsorption and the molecular interaction energy between adsorbates. It is found that mean relative deviations between the experimental results and those predicted by L-F equation are less than 1%, and an adsorbent having a larger specific surface area has a higher adsorption amount and a bigger isosteric heat of adsorption. It is also noted that molecular energy among methane molecules within the adsorption layer on the carbon black is the largest in comparing with those on the GS and the activated carbon. Results also reveal that interaction mechanism between methane and the activated carbon is similar to that between methane and the GS. It suggests that the GS-related storage medium for ANG should have a higher specific surface area.

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1. Introduction

Due to the possibility of obtaining a high energy storage density at ambient temperature and pressure about 3.5–4 MPa, ANG has been being considered as a promising technology since 1980s [1–3]. A large amount of researches have been conducted to develop high performance adsorbents, which are believed to be the base for the ANG technique, and the activated carbon having high specific surface area and micro-porous volume is considered as a suitable medium for natural gas storage [4]. Recently, the US Department of Energy ARPA-E program announced the MOVE (Methane Opportunities for Vehicular Energy) projects, and the

targeted storage density of ANG systems was reset at 12 MJ/kg, which is equivalent to 9.2 MJ/L for fuel volumetric density [5]. These means that development of a new kind of the adsorbent is still a necessity since the storage density on currently available activated carbons cannot meet the targets.

It is believed that the adsorption performance of an adsorbent is closely correlated with the state of the adsorbate confined within the pore, and can be described by the interaction energy among adsorbates and that between adsorbent and adsorbate [6]. Therefore, optimization of gas storage capacity of adsorbents for ANG should be based on those adsorbents that can exert a stronger interaction potential on the adsorbate. Recently, some researches reveal that, although the graphene-related adsorbents had smaller specific surface areas and pore volumes, these adsorbents had larger isosteric heats of adsorption than the activated carbon for such

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gases as methane, carbon dioxide and hydrogen [7–10]. In consideration of that the specific surface areas of the tested graphene-related adsorbents are smaller than $1000 \text{ m}^2/\text{g}$ and that of the theoretic limit of a single-layered graphene is $2630 \text{ m}^2/\text{g}$, increasing the storage density of natural gas on the graphene-related material by enlarging the specific surface area is therefore a possibility [9,11]. Moreover, due to two dimensional layered structure and serried honeycomb crystal lattice of graphene materials composing SP^2 bonded carbon atoms, it is noted that gas adsorption on graphene materials can be more easily enhanced by doping organic ligands or loading metal ions on the surface defects than other adsorbents such as zeolite, activated carbon and metal organic frameworks [10–14]. Hence, analysis of adsorption equilibrium by the theoretic approach is necessary for probing into the reason for the difference between natural gas adsorption on above adsorbents.

Generally speaking, analysis of adsorption equilibrium of natural gas on the adsorbents can be performed by macrothermodynamics approach or through statistical mechanics, and the former is represented by adsorption models while the later is mainly from molecular simulations [15,16]. Due to the high accuracy in predicting the adsorption data, application of adsorption models on analyzing the adsorption equilibrium is an important approach to the system design [6,17–19]. However, the values for parameters of models concerned are needed to be defined by adsorption data, and the results from the models cannot supply information about the state of adsorbed adsorbate directly [17,20]. Molecular simulations and density functional theory calculations do give information about the state of adsorbate, but the computational process is troublesome and time-consuming and the accuracies of the results depend on the pre-assumption of pore structure and pore size distribution (PSD) of adsorbents [16,21–23]. Hence, other theoretical approaches are required to

develop for directly correlating the adsorption data with the state of adsorbed adsorbate molecules. Theoretically, in the viewpoint of thermodynamics, the adsorption equilibrium state of natural gas on the adsorbent should be in energy equilibrium, and energy balance of the equilibrium state should be an effective way of determining molecular interaction energies [6]. These have been verified by the conclusions drawn in relevant researches, which shows that the lattice theory based approximate models can much conveniently determine the interaction energy among adsorbate directly from adsorption equilibrium data [24–26].

In this paper, analysis of adsorption equilibrium of methane on activated carbon, carbon black and GS is carried out in terms of the value of the isosteric heat of adsorption and the interaction energy among adsorbates and between adsorbents. The methane uptake was determined by volumetrically measuring the adsorption isotherms. The absolute adsorption amounts were calculated by L-F equation and then used to calculate the isosteric heat of methane adsorption. A lattice theory based approximate adsorption model was finally employed to calculate the interaction energy among adsorbed methane molecules.

2. Experimental

2.1. Structural characterization

The GS sample was prepared from the thermally expanded r-GO which was synthesized by modified Hummer method. A kind of coconut shell activated carbon AC-01 was co-developed by Ningde Xinsen Activated Carbon Ltd, and graphitized carbon black BP280 was supplied by Cabot Corporation.

Adsorption data of argon at 87 K measured by employing Micromeritics ASAP2020 are presented in Fig. 1. The results from

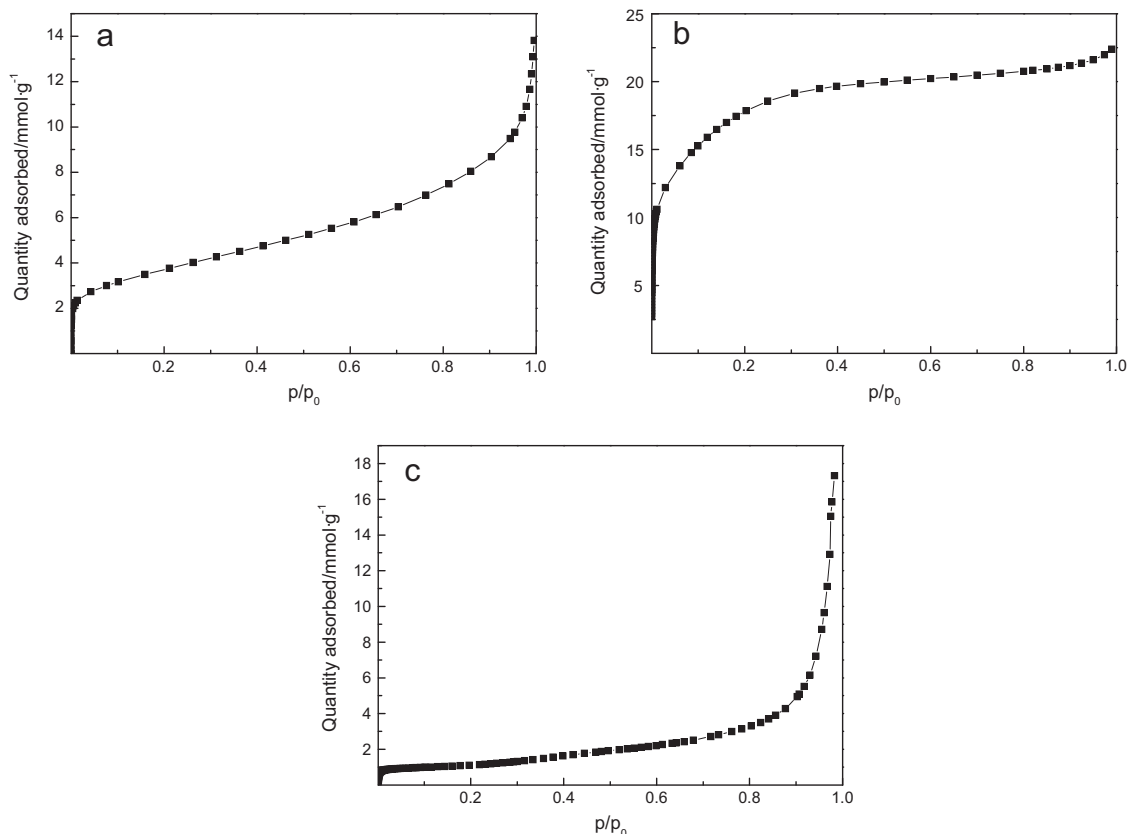


Fig. 1. Adsorption isotherms of argon at 87 K on GS (a), activated carbon AC-01 (b) and carbon black BP280 (c).

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