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ScienceDirect

Procedia Engineering 148 (2016) 600 – 608

Procedia Engineering

www.elsevier.com/locate/procedia

4th International Conference on Process Engineering and Advanced Materials

Description of Carbon Dioxide Adsorption and Desorption onto Malaysian Coals under Subcritical Condition

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Abstract

Coal bed seams have been considered as promising sequestration reservoirs for CO_2 disposal to mitigate the green house gas emissions. The CO_2 adsorption and desorption attributes of CO_2 on dry Malaysian coals (Sarawak, volatile bituminous) were performed using a sorptomat apparatus (ASAP 2010, Micromeritics, USA) and BELSORP-mini II machine (BEL Japan, Inc.) at 273 K, 298 K and pressure up to 1 bar. The CO_2 adsorption was favourable at low temperature and dry coal conditions. However, S3 and S4 coals have the highest adsorption capacity by 0.71 and 0.73 mmol/g respectively. According to IUPAC classification of adsorption isotherms, CO_2 adsorption isotherm of all coal samples follow type I which most probably describe the adsorption limited to a few molecular layers (micropores). The results of adsorption and desorption isotherm demonstrate a positive hysteresis in all coal samples. The S1 coal and S2 coal have the highest hysteresis between adsorption and desorption isotherm compared to S3 coal and S4 coal. According to hysteresis classifications, the hysteresis during CO_2 adsorption and desorption process for all coal samples follows type H_3 which describes micropores and mesopores. The evaluation of the equilibrium adsorption data where fitted using by Langmuir, Freundlich, Redlich-Peterson, Koble-Corrigan, Toth and Sips models. Toth model provided the best fit for all adsorption experimental data that predicting all coals having heterogeneous surface properties.

Peer-review under responsibility of the organizing committee of ICPEAM 2016

Keywords: CO_2 adsorption and desorption; coal; hystersis; isother medols.

1. Introduction

The excessive emission of carbon dioxide (CO_2) into the atmosphere is broadly agreed as one of the major causes of global warming and air pollution. The most successful disposal techniques of the captured carbon dioxide

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Peer-review under responsibility of the organizing committee of ICPEAM 2016
doi:10.1016/j.proeng.2016.06.521

are geologic storage sites such as depleted oil and gas reservoirs, deep saline aquifers and deep unminable coal bed seams. Geosequestration of carbon dioxide (CO_2) in coal bed seams is reported as a promising CO_2 disposal option with or without methane displacement process. However, CO_2 sequestration into coal bed seams is combination between physical and chemical adsorption with other trapping processes, mostly due to the heterogeneous morphology of coal texture [1] and extreme coal bed seam conditions.

The storing mechanism of CO_2 in coal bed seams mostly relies on the adsorption characteristics of the morphology and the porous coal structures [2, 3]. According to White et al which reported that 95-98% CO_2 stored by adsorption mechanism in the coal matrix depends on the gas in the coal bed seam and there are other storing mechanisms such as gas stored within the coal matrix composition, free gas and gas stored as a solute in the water porous [4].

Thus, it is essential to comprehend the CO_2 adsorption onto Malaysian coals during sequestration process and the CO_2 desorption mechanism from coal by depressurization process. However, the deviation occurs between adsorption and desorption isotherm of gases on porous materials is called hysteresis. Hysteresis indicates that coal desorbs less gas than the sorption volume in the adsorption isotherm at the same pressure. The hysteresis level is independent of the coal physical properties (density, diffusivity, viscosity, surface texture, and gas phase) [2] and operating condition (pressure and temperature). CO_2 sorption hysteresis process is a favourable for the long-range preserve of CO_2 sequestration [5]. Currently, numerous research studies of CO_2 sorption on various coal specimens have been conducted not just only for practical evaluation of coal bed gas capacity but also to comprehend the fundamental mechanisms of the gas adsorption and desorption isotherms on coal. To understand the interactions of CO_2 with coal, it is necessary to conduct experiment on CO_2 adsorption and desorption isotherm on Malaysian coal from low to high temperatures and pressures.

In this study, the CO_2 adsorption/desorption on dry Malaysian coal measured under subcritical conditions. The CO_2 adsorption isotherm conducted at 273 K, 298 K and pressure up to 1 bar. Meanwhile, desorption was performed by depressurizing CO_2 from 1 bar. The results from two different temperatures were compared to each other. Finally, the adsorption isotherm models were utilized to fit the equilibrium adsorption data.

2. Experimental

2.1. Adsorbent

The coal samples were obtained from Merit-Pila coal mine Lower (S1), and Upper (S2) zones and Mukah-Balingian coal mine from Area1 (S3) and Area4 (S4) coordinates, Sarawak, Malaysia. The majority of coals in Sarawak are sub-bituminous coals. Before adsorption measurements on dry coal, the samples were desiccated for more than 12 hr in a vacuum oven (pressure ~ 10kPa) at 378K (105 °C).

2.2. Instruments and Experimental Procedures

Experimental studies of sorption were firstly performed with the use of Sorptomat instrument, ASAP 2010 (Micrometrics, USA) to study CO_2 adsorption and desorption of S1, S2, S3 and S4 coals. CO_2 (99.99%) adsorption conducted at 273 K (0°C) at 760 mmHg (1 bar). At each target pressures, the gas is stored inside the manifold (reservoir) before release onto sample. The pressure was monitor before/after the stored gas in the manifold is release onto sample. When adsorption occurs, the pressure starts decline. By using the gas law the volume of adsorbed is determined by determined the changes in pressures while the temperature is held constant. The volume adsorbed is finally calculated after minus the void volume inside the sample tubes.

For carbon dioxide gas (CO_2), degas was performed to clean the surface of the coal samples prior to analysis. The samples were subjected to constant nitrogen purging while heating the samples to remove any sorbed species from the sample. For the temperature profile of degas, the first hour the temperature was heated up to 90°C to remove the moistures for 60 minutes (1 hr). This is followed by raising the temperature up to 150°C at 10°C/min for 480 minutes (8 hrs) to remove any sorbed species from the samples. Eventually, the samples were subjected to CO_2 gas adsorption using the bath temperature of 0°C by controlled water circulating bath. A target pressure is gradually increased for up to about 800 mmHg and each target pressure points, the amount of CO_2 adsorbed was