



Full Length Article

The temperature effect on the methane and CO₂ adsorption capacities of Illinois coal



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ABSTRACT

The sorption capacity of coal is influenced by temperature, coal rank, mineral composition and moisture content. In the study, we experimentally measure the methane and carbon dioxide sorption capacity at various temperature for Illinois coal. A volumetric adsorption apparatus was employed to determine the excess gas adsorption capacity, and then the absolute adsorption capacity was estimated through the adsorbed gas phase density correction at each tested pressure. The adsorption isotherms were estimated for various temperatures from 283.15 K to 343.15 K. The experimental results indicated that both the methane and carbon dioxide adsorption capacities decrease linearly with an increase in temperature from 283.15 K up to 323.15 K, and the adsorption capacity reduction for CO₂ is more significant than that for methane. When the temperature is equal to or greater than 323.15 K, the adsorption capacities are independent of the temperature and remain constant for both gases. This means that there is a “critical temperature point” at which the influence of the temperature on the adsorption capacity vanishes, and the adsorption isotherms are very similar when the temperature is beyond the critical temperature point. The ratios of the CO₂/CH₄ absolute adsorption capacities vary from 2.62 and 3.80 with the tested pressures and temperatures.

1. Introduction

Coalbed methane (CBM) is one of the most important unconventional natural gas resources. Both the gas production and the estimated CBM reserve have increased sharply during the last decades due to the advanced extraction technologies. The most recent data from the Energy Information Administration (EIA) states that the proven CBM reserves of the USA accounted for approximately 17.5 trillion cubic feet (TCF) in 2010. The CBM production in 2010 was 1.9 TCF, which was approximately 8.5% of the US natural gas production [1–3]. In addition to the primary CBM production, the CO₂ sequestration in unmineable coal seams has drawn significant attention in last decade since it not only can potentially sequester the CO₂ in a relatively low reservoir pressure as an adsorbed gas but can also effectively enhance the methane production by CO₂ displacement and sweeping [2,4,5]. For both the primary gas production and CO₂-sequestration in the CBM reservoirs, understanding the adsorption behavior of the CH₄ and CO₂ in the coal is of great importance, and the storage capacity of the gases will be the first important parameter for the coal reservoir evaluation.

The methane retained in a coal reservoir is a function of the reservoir pressure, temperature, maceral composition, mineral content,

and pore microstructures. Thus, the effects of these factors on the adsorption capacity have been thoroughly investigated for the past a few decades. Among these factors, the temperature is one of the most important factors influencing the amount of gas adsorbed in a CBM reservoir. The temperature in the coal seams varies with the depth and usually has an increasing trend with the increase of the burial depth [6–9]. The virgin reservoir temperature at a given depth h can be estimated by the geothermal gradient multiplied by h and added to the ground temperature [10]. As is commonly used, the geothermal gradient is 1.8 K/100 m at a relatively shallow depth and the ground temperature used is 284.15 K. The temperature at depth is estimated by the following relationship:

$$T = 1.8 \frac{h}{100} + 284.15 \quad (1)$$

In addition to the geological influence on the temperature in the CBM reservoirs, the effects of the change in the temperature are much more significant in a scenario of CO₂-sequestration, which is associated with enhanced-CBM (ECBM) production. Since 1993, several particular sites in San Juan Basin where CO₂ was injected for sequestration and ECBM have been set up for pilot experiments [4,11,12]. The injection process and storage of the CO₂ in the subsurface systems are briefly

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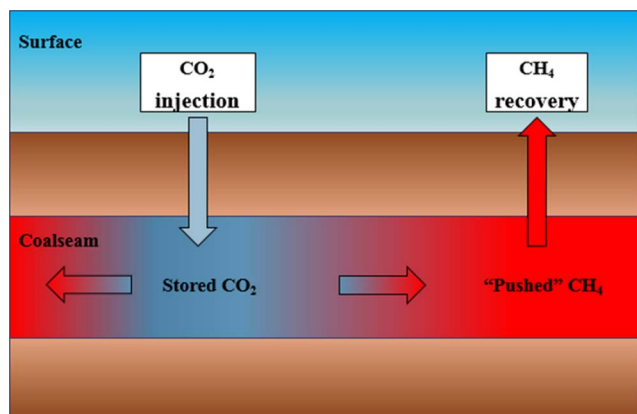


Fig. 1. The mechanism of CO₂ storage and ECBM underground [13–15]. The blue and red sections represent low-temperature and high-temperature regions in an underground coal seam, respectively.

summarized and illustrated in Fig. 1.

The mechanism by which CO₂ can enhance the CBM recovery, along with the CO₂ storage, is a complex mix of physical and chemical interactions that must achieve equilibrium simultaneously in the sorbed state and in the gaseous state [4]. It was commonly known that the equilibrium ratio of CO₂ to CH₄ in the adsorbed state was greater than one. When CO₂ is injected into the coal seam, the partial-pressure of CH₄ in the gaseous phase within the cleat system is reduced instantly, and the CO₂ molecules are preferentially adsorbed onto the coal, displacing the CH₄ molecules. Based on the two processes occurring via the CO₂ injection, CO₂ remains in the system and the trapped methane is “pushed” off from the coal matrix by the CO₂.

In a continuous CO₂ injection, the coal seam temperature can vary with time due to the CO₂ phase changes [16–18]. Practically, before the CO₂ injection, the CO₂ is condensed to a nearly liquid stage with an extremely low temperature of approximately 236.15 K and a density of 1.1 g/cm³. The relationships between the pressure/temperature and density/pressure are shown in Fig. 2. When the condensed CO₂ enters the atmospheric pressure, it will gasify instantly. During the rapid vaporization of the liquid CO₂, the gas will absorb much heat, nearly 578 kJ/kg, from the ambient environment, which can play a significant role in cooling a certain area around the injection well. When it is applied to a subsurface injection, e.g., at a depth of more than 750 m, where the temperature is approximately 313.15 K or more and the pressure is more than 7 MPa [14], the temperature of the coal will

decrease to ~283.15 K under the cold CO₂ injection condition. If the CO₂ injection ceases, the temperature will rebound back to the *in situ* temperature because of the heat transferred from other parts of the reservoir. Therefore, the temperature of the coal seam varies with time under the CO₂ injection condition, which will automatically change the gas adsorption capacity at different temperatures.

The gas adsorption capacity of the coal shows differences in the temperature evolution process. The phenomenon that the adsorption capacity of gas, including methane and carbon dioxide, has an obvious decrease as the temperature increases has been indicated by many studies, which are detailed and reviewed in the next section.

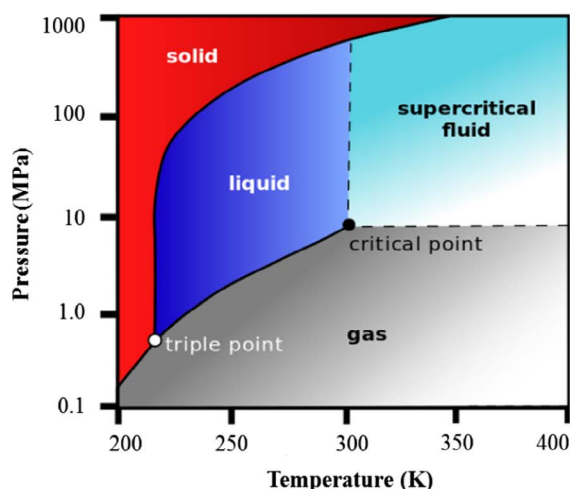
2. Background and literature review

2.1. The adsorption capacities of CH₄ and CO₂ in coal

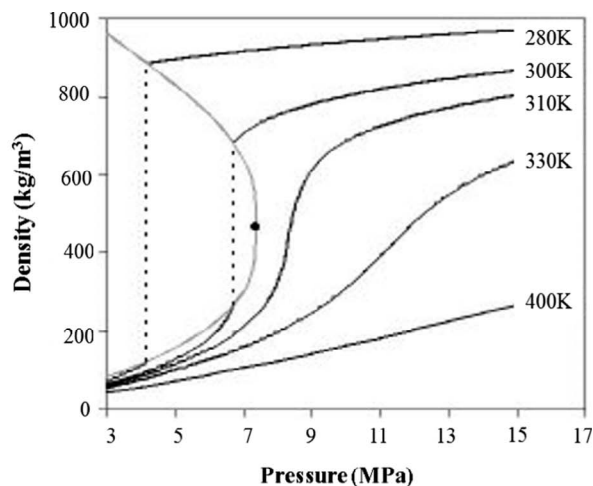
The gas adsorption process in coal is complex because of the unique characteristics of the coal's pore structure. Coal is known to be rich in micropores, which provide extensive internal surface areas where the gas molecules will reside. During recent years, the issues of CO₂ storage and enhanced coalbed methane recovery have been addressed in numerous publications [15,21–23]. CO₂ and CH₄ adsorb competitively on the coal surface, and an understanding of each gas individually is important to make a comparison and predict the feasibility of the approach for permanent CO₂ storage. Many of the publications show how CO₂ adsorbs in relation to CH₄, with a ratio of 2:1, due to the higher adsorption energy and smaller molecular diameter of CO₂ [24–26]. However, there are also some studies indicating that the adsorption ratio varies more widely, from 10:1 for low ranking coals to less than 2:1 for low to medium volatile bituminous coals [27–29].

2.2. Coal-gas sorption modeling

There have been several sorption isotherm models proposed to define the adsorption process of a gas in porous solids. For a gas-coal system, a few popular models have been applied to model the gas sorption behavior in coal, including Langmuir, BET, Dubinin-Astakhov (D-A), Dubinin-Radushkevich (D-R), and simplified local-density (SLD) models [29,30]. Among these models, the Langmuir model is the most commonly used model due to its simplicity and reasonable accuracy [31,32].



(a) CO₂ pressure-temperature relationship



(b) CO₂ density-pressure relationship

Fig. 2. CO₂ pressure-temperature & density-pressure phase diagram [19,20].

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