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Full Length Article

Pore variation of three different metamorphic coals by multiple freezing-thawing cycles of liquid CO_2 injection for coalbed methane recovery

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ABSTRACT

Liquid CO_2 (LCO_2) enhanced coalbed methane recovery had been studied in laboratory experiments and field applications, supporting many improvements and achievements. Previous studies primarily investigated the gas bursting, flooding effect and adsorption effect; however, the freezing-thawing phenomenon (drikold formation and gasification) that commonly occurs during the LCO_2 injection process was insufficiently studied. The freezing-thawing phenomenon might enhance the pore volume and change the permeability evolution of the coalbed; thus, cyclical LCO_2 injection was proposed to exploit the phenomenon, and the influence of multiple freezing-thawing cycles on the coal pores was investigated in this paper. Nuclear magnetic resonance (NMR) and infrared thermal imagery (ITI) were used to monitor the pore variation and surface temperature distribution, respectively. Low temperatures could make the saturated water in the pores freeze and undergo a 9% volume increase. The three coals used in this experiment displayed different crack intensities and forms with ITI. After cyclical LCO_2 injection, the NMR amplitude increased, and the T_2 range was widened under a saturation condition, while the cores under a centrifuge state had lower amplitudes and a narrower T_2 range; this difference indicated that the pore structure could be altered by multiple freezing-thawing cycles of LCO_2. The more freezing-thawing cycles the cores experienced, the greater the change in pore structure was. The total porosity φ_t and effective porosity φ_e increased while the residual porosity φ_r and $T_{2cutoff}$ values decreased with more freezing-thawing cycles. However, the variations with coal rank were observed; with higher coal ranking, φ_t and φ_e increased less, and the φ_r and $T_{2cutoff}$ values decreased less, which suggests that lower ranking coals could be most easily affected by the LCO_2 enhanced recovery method and have the most improved pore connectivity. Moreover, the enhancement ratio of φ_t and φ_e increased for all three coals tested, which could be fit with quadratic functions with fit coefficients greater than 0.99. The increasing relative ratio $D_{e/t}$ of anthracite was fit with a linear function, while the lignite and bitumite were fit with quadratic functions. These functions all indicate that the multiple freezing-thawing cycles of LCO_2 injection had a positive impact on the enhancement efficiency of pore porosity. Finally, a potential field application of cyclical LCO_2 injection was also discussed to improve the fracturing effect.

1. Introduction

Coalbed methane (CBM), due to its efficient clean-burning and high calorific value, has become a promising alternative new resource and is attracting more interest from government, scholars, and entrepreneurs [1–4]. Statistically, in China, the CBM resources buried below a depth of 2000 m has a volume of 36 trillion m^3, and mines with over 10 m^3/t gas content account for 41% of the Chinese CBM reserves [5,6]. However, the permeability of most coal mines are 10^{-4}–10^{-2} mD, which is three orders of magnitude lower than that of the United States [7–9].

Enhanced CBM recovery (ECBM) by injecting carbon dioxide (CO_2) or nitrogen (N_2) in a gas or liquid phase, is a method of

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waterless fracturing to recover CBM and has advantages such as no water-blockage, no water-sensitivity, and no residues [10,11]. Compared to N_2, CO_2 has a larger adsorption capacity than methane (CH_4), which enhances CO_2 flooding during the injection and storage process [12,13]. CO_2 has acidification characteristics that could remove blockages in the pores and fissures [14]. In addition, the N_2 separation from the produced CBM gas will add extra costs. In the Asia-Pacific, the CO_2 content in the gas can be up to 20% at the point of sale [15].

The first patent of liquid CO_2 (LCO_2) fracturing technology was registered by Bullen and Lillies [16]. The field testing of LCO_2 fracturing technology was by trial and error over the past several decades [17–22]. Earlier studies focused on technical designs [23,24], performance of the LCO_2 fracturing system [25–28], the equipment and technology [29], prospects and development trends of LCO_2 fracturing [30], as well as other aspects. Many pilot tests had been conducted in countries such as the USA, Canada, China, the Netherlands, Italy, and Japan [31–33]. A CO_2-ECBM pilot test was carried out in 2004 by China United CBM Co. Ltd. in collaboration with the Alberta Research Council from Canada, which was designed with a 5-spot pattern around the injection well with an area of 0.6 km^2; it was reported to be a considerable enhancement to the CBM recovery [34]. In 2015, LCO_2 with sand fracturing was successfully implemented in the Shenmu gas field, with a total volume of LCO_2 and ceramsite sand of 355 m^3 and 9.6 m^3, respectively. After fracturing, the well produced industrial gas at 4.2×10^4 m^3 per day.

Earlier studies mainly focused on a single injection and large volume of LCO_2 to achieve fracturing by gas bursting, in both field applications and laboratory experiments. However, the phenomenon of freezing-thawing in the local coal due to the formation of temperature gradient [35], and the cryogenic effect of LCO_2 was generally neglected. Some studies [36–38] verified that the multiple freezing-thawing cycles could weaken the coal strength and produce many new cracks. How to make use of the freezing-thawing effect generated during LCO_2 injection is a relatively new research topic in need of comprehensive investigation before cyclical LCO_2 injection is expected to be applied in the field, in a similar way as pulse hydraulic fracturing [39–41].

In this paper, the influence of multiple freezing-thawing cycles on the pore system from LCO_2 injection was investigated. Three metamorphic coals (lignite, bitumite, and anthracite) were selected and cored for this study. An innovative cryogenic loading system was established to study the degradation behavior of the coals. Infrared thermal imaging (ITI) was used to measure the temperature distribution of the core surfaces. Nuclear magnetic resonance (NMR) was used to investigate the pore and porosity changes of the three cores by analyzing their transverse relaxation (T_2) distributions, and the relationships between the freezing-thawing cycles and porosity change or enhancement efficiency were assessed. Finally, the potential application of cyclical LCO_2 injection in CBM recovery was discussed.

2. Experimental materials, equipment and procedures

2.1. Preparation of coal cores

Coal cores were prepared from the Shengli Coal Mine, Inner Mongolia (IM-1), the Datong Coal Mine, Shanxi (SX-1), and the Yangzhuang Coal Mine, Huaibei (HB-a), and their locations are shown in Fig. 1. These cores were drilled with a diameter of 50 mm and height of 60 mm, and satisfied the following precision requirements: non-parallelism error and non-flatness error of the two end-faces less than 0.005 mm and 0.002 mm, respectively, diameter error along the sample height of no more than 0.3 mm, and a maximum error of end-face verticality perpendicular to the sample axis less than 0.25°. All the cores were wrapped in preservative film and put in a core-curing room for preservation. The proximate analysis and maceral analysis of the three cores are listed in Table 1.

2.2. Experimental system and equipment

An innovative cryogenic loading system was created, as shown in Fig. 2. The apparatus included three main subsystems: hydraulic loading equipment, monitoring equipment, and LCO_2 injection equipment. The hydraulic loading system could provide uniaxial compression (0–18 MPa), and the cores were placed in a sealed chamber that was wrapped with thermal insulation material. Pressure of more than 0.5 MPa was be relieved by a decompression safety valve. Monitoring equipment consisted of thermocouple (HY101) and pressure sensor (CY200), which measured the temperature and pressure of CO_2 gas in the chamber, respectively, in turn they further qualitatively represented the environment temperature around the cores during the LCO_2 gasification process and determined the injection cycles (number of constant pressure

Fig. 1. Location of the three coal cores tested in this study.