



Contents lists available at ScienceDirect

International Journal of Rock Mechanics and Mining Sciences

journal homepage: www.elsevier.com/locate/ijrmms

Experimental investigation of the effects of supercritical carbon dioxide on fracture toughness of bituminous coals



Jianfeng Yang^{a,b}, Haojie Lian^{a,b}, Weiguo Liang^{a,b,*}, Vinh Phu Nguyen^c, Yuedu Chen^{a,b}

^a College of Mining Engineering, Taiyuan University of Technology, Taiyuan, Shanxi, China

^b Key Lab of In-situ Property-improving Mining of Ministry of Education, Taiyuan, Shanxi, China

^c Department of Civil Engineering, Monash University, Clayton, Victoria 3800, Australia

ARTICLE INFO

Keywords:

Coal
Fracture toughness
Supercritical carbon dioxide
Soaking time
Modified maximum tangential stress criterion

ABSTRACT

Effects of supercritical carbon dioxide (scCO₂) on fracture toughness of bituminous coals are studied. The semi-circular specimens under three-point bending (SCB) were adopted for conducting experiments on the coal samples after being immersed in scCO₂ for different soaking periods. To investigate the fracture toughness in different loading conditions, including pure mode I, pure mode II, and mixed mode I/II, the inclination angles of the prefabricated cracks in the coal SCB samples were set as 0°, 5°, 15°, 30°, 45° and 54°, respectively. The experimental results show that after being soaked in scCO₂ for 7 days, 14 days and 21 days, the pure mode I fracture toughness of the coals reduced by up to 30.8%, 69.2% and 82.1%, respectively, and the mode II fracture toughness 29.1%, 66.7% and 79.9%, respectively. The fracture toughness in pure mode I loading is more sensitive to the effect of scCO₂ than that in pure mode II loading. The values of the effective fracture toughness K_{eff} in mixed mode I/II loading also declined due to the effect of scCO₂, and the samples with different crack inclination angles exhibited no obvious differences in the degradation degrees of K_{eff} . With increasing soaking time, the compaction stage extended, and the slope of the load–displacement curve in the elastic stage decreased significantly. The microcracks on the surface of the coal SCB specimens gradually increased with the soaking time increasing. In addition to causing the reduction of surface energy and swelling of coals, scCO₂ mobilizes organic molecules from coals, leading to the modification of the physical structure of the coal matrix and the degradation of fracture toughness in various loading modes. Based on the modified maximum tangential stress (MMTS) criterion, which takes into account the effects of T-stress, the theoretical values are in better agreement with experimental results compared with the traditional maximum tangential stress (MTS) criterion.

1. Introduction

Storage of CO₂ in unmineable coal seams is regarded as a technology with huge potential to reduce greenhouse gas emissions.^{1–3} In addition, since the adsorption capacity of CO₂ is higher than CH₄, injection of CO₂ into coal seams can displace the coalbed methane (CBM) and thus enhance the CBM exploitation.^{4,5} When CO₂ sequestration takes place at deep coal seams, where the pressure can exceed 7.38 MPa and temperature might exceed 31.1 °C, the CO₂ reaches the supercritical state. Fig. 1 is the phase transition diagram of CO₂, from which it can be seen that the critical conditions for CO₂ phase transition can be satisfied in the deep geological environment. Supercritical carbon dioxide (scCO₂) has distinctive physical-chemical properties, such as low viscosity like a gas and high density like a liquid.^{6–9} For this reason, scCO₂ is also considered as an ideal non-aqueous fracturing fluid which can produce denser crack network.¹⁰

The adsorption of scCO₂ can influence the physical and mechanical properties of rock materials,^{11–13} which further affects the rock stability and CO₂ sequestration process.¹⁴ According to Vishal and Singh¹⁵ and Perera et al.,⁹ the coal permeability declines rapidly on account of the effect of scCO₂. Day et al.¹⁶ observed that scCO₂ can lead to the volumetric swelling of coals by an optical method. Perera et al.¹⁷ carried out a uniaxial compression experiment on bituminous coals after the reaction with scCO₂, and found that the uniaxial compressive strength (UCS) of the coals reduces by 79% and the elasticity modulus (E) by 74%, and the rate of decrease is larger than that of the coals after reaction with the gaseous CO₂ (gaseous CO₂ saturation was observed to reduce UCS of coals by 53% and E by 36%). Ranathunga et al.¹⁸ drew the similar conclusion for low rank coals. More importantly, the interaction between scCO₂ and coals may trigger the initiation of cracks or propagation of existing cracks under in-situ stresses, which has an enormous impact on the CO₂ storage and scCO₂ fracturing. To the best

* Corresponding author at: College of Mining Engineering, Taiyuan University of Technology, Taiyuan, Shanxi, China.
E-mail address: master_lwg@hotmail.com (W. Liang).

<https://doi.org/10.1016/j.ijrmms.2018.04.033>

Received 10 January 2018; Received in revised form 26 March 2018; Accepted 19 April 2018

Available online 29 May 2018

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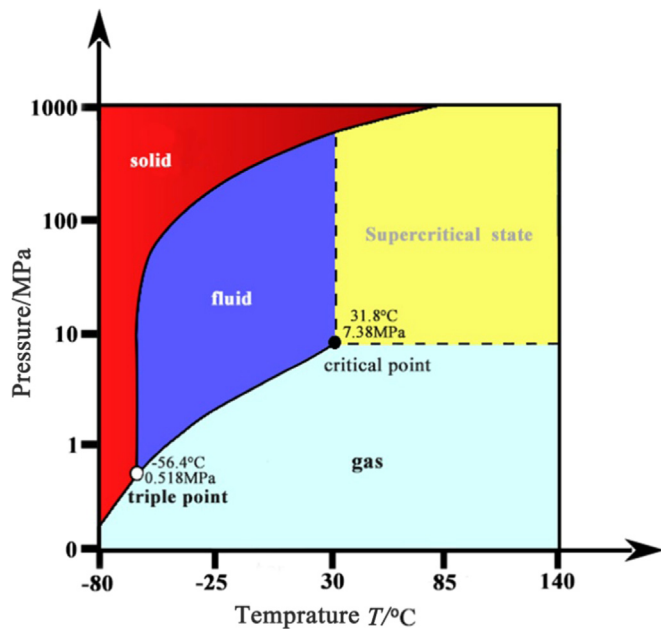


Fig. 1. The phase transition diagram of carbon dioxide.

of the authors' knowledge, there is still a lack of research on the effect of scCO₂ on crack propagation in coals. This paper is intended to fill this research gap.

In the framework of linear elastic fracture mechanics (LEFM), Irwin¹⁹ defined the stress intensity factor (SIF) to describe the stress fields in the neighborhood of crack tips. The critical value of SIF corresponding to crack propagation is referred to as the fracture toughness, which characterizes the resistance of materials against crack propagation. Bhagat²⁰ experimentally studied the mode I fracture toughness of coals and derived the relationship between the mode I fracture toughness and the tensile strength of coals. Zipf and Bieniawski²¹ researched the fracture toughness of coals in mixed-mode loading. Zhao et al.²² investigated the dynamic fracture toughness of coals. The mode I fracture toughness of the coals with bedding structure was investigated by Wu et al.²³.

In this research, we used a semi-circular specimen under three-point bending (SCB) to test the fracture toughness of coals for different soaking time in scCO₂ in pure mode I, pure mode II and mixed-mode I/II loading. Furthermore, the experimental results are compared with the theoretical values based on the modified maximum tangential stress criterion.

2. Experimental methods and progress

2.1. The SCB specimen under three-point bending

To study the fracture toughness of rock materials in the loading conditions varying between pure mode I, pure mode II, and mixed mode I/II, a variety of specimens and experimental methods can be adopted, such as the cracked chevron notched Brazilian disc specimen,²⁴ the four-point shear specimen,²⁵ the compact tension-shear specimen²⁶ and the semi-circular specimen under three-point bending (SCB).²⁷ Among them, SCB is a particularly favoured experimental configuration for testing fracture toughness (including mode I, mode II and mixed-mode I/II) for its simplicity of machining and testing process, so SCB is adopted in this experiment. Chong et al.^{28–30} put forward to use SCB specimens to test the diverse mode fracture toughness of rocks. Lim et al.³¹ simulated the SIFs of SCB specimens for a wide range of specimen geometries and demonstrated that the SCB technique is applicable to testing fracture toughness from mode I to mode II.

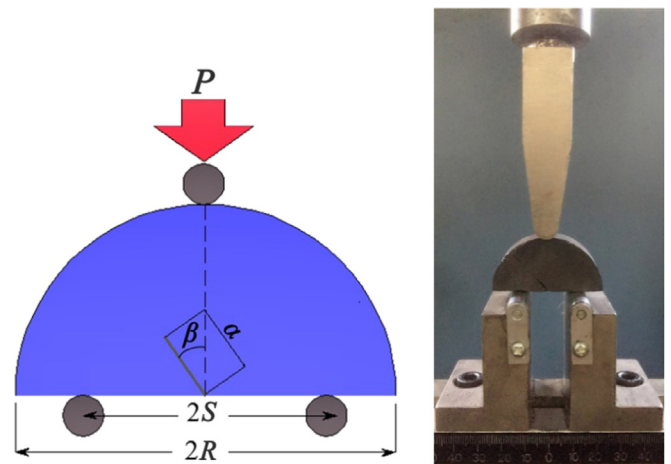


Fig. 2. The SCB specimen.

As shown in Fig. 2, the specimen in SCB test is a semi-circular disc with a radius R , and from its center a radial edge crack with length α is prefabricated. Fracture toughness experiments are performed by exerting a vertical compressive force (P) along the axis of the specimen under the three-point-bending apparatus. The mode I and mode II SIFs for the SCB specimen are denoted by K_I and K_{II} , and the critical values of K_I and K_{II} at which the prefabricated crack will propagate are Mode I fracture toughness K_{Ic} and mode II fracture toughness K_{IIc} , respectively. The mode I and mode II SIFs for the SCB specimen are given by Ayatollahi and Aliha³² as:

$$K_I = \frac{P\sqrt{\pi\alpha}}{2RB} Y_I \left(\frac{\alpha}{R}, \frac{S}{R}, \beta \right) \quad (1)$$

$$K_{II} = \frac{P\sqrt{\pi\alpha}}{2RB} Y_{II} \left(\frac{\alpha}{R}, \frac{S}{R}, \beta \right) \quad (2)$$

where P is the compressive force, β is the angle by which the pre-fabricated crack line departs from the vertical direction, B is the thickness of the specimen, α/R is the ratio of the crack length to the semi-circular radius, and S/R is the ratio of the half distance between two supporting cylindrical rollers to the radius. Y_I and Y_{II} are defined as the mode I and mode II geometry factors, respectively, which are functions of the geometric parameters β , α/R and S/R . When the compressive force P reaches the critical value (the peak load), P_{cr} , the crack starts to propagate, and the values calculated using Eqs. (1) and (2) are fracture toughness K_{Ic} and K_{IIc} , respectively. Different modes of fracture toughness can be obtained by changing the geometric parameters. For example, if β is set to be zero, then the SCB specimen is subjected to pure mode I loading. Ayatollahi and Aliha³² calculated the values of Y_I and Y_{II} corresponding to pure mode I and pure mode II loading conditions for different geometric factors.

2.2. Specimen preparation and experimental procedures

The bituminous coals used in this experiment were taken from a depth between 350 m and 400 m in Linfen City, Shanxi Province, China. The proximate analysis result of this kind of coals is shown in Table 1. The cylindrical coal samples with a diameter 50 mm were obtained using a 1.5 mm carborundum wire saw in a numerical control machine tool, in which way the risk of collateral machining damage of the coal samples can be reduced greatly. The cylindrical specimens were then cut to be disc coal specimens with 20 mm thickness, which were further cut into two equal semi-circles. For each semi-circle a straight notch was prefabricated by using a 0.5 mm diamond-impregnated fine wire saw, and the ratio of the crack length to the semi-circular radius (α/R) was 0.35. In order to arrive at multiple loading modes, the angle (β) by which the crack line departed from the symmetric line of the semi-circle

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