

Experiment study of mechanical properties and microstructures of bituminous coals influenced by supercritical carbon dioxide

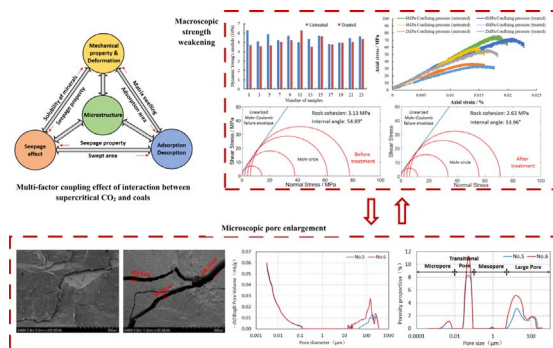
Meng Meng^{a,*}, Zhengsong Qiu^b

^a Department of Petroleum Engineering, The University of Tulsa, Tulsa, OK, USA

^b Institute of Petroleum Engineering, China University of Petroleum (East China), Qingdao, China



GRAPHICAL ABSTRACT



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ABSTRACT

The injection of CO₂ into deep coal seams can not only increase the recovery of CH₄ but also contribute to the geological sequestration of CO₂. In deep coal seams, CO₂ can easily become the supercritical state when the pressure is over 7.38 MPa, and the temperature is over 31.04 °C. It can influence both the physical and chemical properties of coals, especially weakening the mechanical strength, which could compromise the long-term integrity and stability of the deep coal seams. Through acoustic emission experiment and triaxial compression experiment, the results show that after the treatment of supercritical CO₂, the mechanical parameters of coals, including dynamic Young's modulus, static Young's modulus, rock cohesion, and peak strength, decrease significantly. It demonstrates that supercritical CO₂ can reduce the mechanical strength of coals. This macroscopic phenomenon can be explained by the mechanism of the enlargement of microscopic pore spaces of coals, and this mechanism has not been studied thoroughly yet. Therefore, several microscopic quantitative experiments are comprehensively conducted, including scanning electron microscope (SEM), mercury injection porosimetry (MIP) and nuclear magnetic resonance (NMR). The results of these three tests are relatively consistent, and they show that after treatment of supercritical CO₂, not only the diffusion space consisting of micropore and transitional pore but also the percolation space involving large pores and even cracks has been enlarged and expanded. This can be a significant underlying microscopic mechanism to effectively explain the weakening of the mechanical strength of coals. The pore space enlargement, together with the swelling of coal matrix, and the theoretical explanation of fracture mechanics and thermodynamic theory are all underlying mechanisms to explain the weakening behavior of coals influenced by the supercritical CO₂.

* Corresponding author at: The University of Tulsa, McDougall School of Petroleum Engineering, North Campus 211D, USA.
 E-mail address: mem282@utulsa.edu (M. Meng).

1. Introduction

The injection of CO₂ into coal seams can not only be used to improve the recovery of coal seams methane but contributes to the geological sequestration of CO₂ [1–3]. The interaction between CO₂ and coal seams is a complex process that consists of several physicochemical phenomena, including the change of microstructure, the adsorption/desorption of CO₂ and CH₄, the seepage effect of CO₂ and CH₄, the deformation of coal matrix and the changing of the mechanical strength of coal seams. These processes are closely related and coupled with each other. When CO₂ is injected and flows in the deep coal seams, it can easily become the supercritical state ($P > 7.38$ MPa, $T > 31.04$ °C). Then it will change the pore pressure and effective stress in the coal seams. Within the affected area, CO₂ can displace CH₄ through competitive adsorption. As time pass by, the pressure of CO₂ will dissipate gradually, and desorption will happen. The desorption of CH₄, adsorption of CO₂ and finally desorption of CO₂ could cause a variance of mechanical properties and volume deformation of coals. The physical parameters of the coal, such as porosity and permeability will also be changed. All these, in turn, will affect the seepage properties of CO₂ in the coal seams. Therefore, we can see that the injection of CO₂ into deep coal seams is a complex process with different mechanisms coupled with each other and the whole process is shown in Fig. 1.

Up till now, many researchers have paid enough attention to the adsorption, seepage, and volume swelling behavior of supercritical CO₂ on coals. However, researchers have not paid enough attention to the variance of mechanical properties and microstructures of coal under the influence of supercritical CO₂. There is only a few literature related to this area, and unfortunately, most of them are limited to the subcritical or gas state of CO₂. Ranjith [4] used acoustic emission methods to study the effect of CO₂ saturation on mechanical properties of coal and showed that the sorption of CO₂ could cause a reduction in strength of the coal samples. But he didn't provide the detailed study of the actual underlying mechanisms. Perera [5] studied the influence of mechanical property of coals influenced by CO₂ and N₂. He concluded that CO₂ saturation could weaken the mechanical strength of coals, but N₂ saturation could slightly increase the mechanical strength and delays crack propagation. Masoudian [6] studied the effect of CO₂ on the mechanical properties of coal under in-situ stress conditions and he suggested that the adsorption of CO₂ changes Young's modulus and the strength of coal sample and these effects are reversible. It shows that the Young's modulus of coal decreases up to 19% and the strength of coal reduced by 20%.

For the influence of supercritical CO₂, Perera [7] used uniaxial compression test to investigate the effects of gaseous and supercritical

CO₂ on coal strength. He concluded that the supercritical CO₂ has a greater influence in weakening coal strength and Young's Modulus. Ranathunga [8] used the unconfined compressive strength tests, acoustic emission system and optical 3D deformation analysis to study the mechanical property alterations of coal. He found that supercritical CO₂ has a greater ability to reduce the coal strength because supercritical CO₂ has a greater adsorptive potential, which eventually creates greater coal matrix swelling. Later, Ranathunga [9] studied the influence of supercritical CO₂ on coal seams during CO₂ adsorption under in-situ stress conditions by conducting tri-axial compression tests. The results show that the strength reduction in coal with the CO₂ injection under field conditions is significantly less than that expected based on uniaxial compression tests.

Among these limited publications, the weakening of the coal strength after its interactions with supercritical CO₂ is a consensus. The macroscopic phenomenon is the overall effect of the microscopic properties, and the variance of microscopic properties can effectively explain macroscopic phenomena. There are three possible mechanisms to explain the weakening effect. First, supercritical CO₂ could expand the pore space of coals. Second, the adsorption of supercritical CO₂ could cause the swelling of the coal matrix [8]. Thirdly, the theory of fracture mechanics and thermodynamic theory could be used to explain the weakening behavior [10]. The last two mechanisms have been studied by former researchers. However, to the best of our knowledge, there is not enough work that thoroughly investigates the microscopic mechanism of pore space enlargement and its relation to the weakening of coal strength. Hol, S. [11–13] analyzed the microstructure of coal under exposure of supercritical CO₂ and mentioned that microfractures formed and propagated through the coal samples as a result of the exposure to CO₂. However, under in-situ conditions adsorption and swelling cannot cause any auto-cracking and the new microfractures should occur during the CO₂ injection process or because of the extraction ability of the supercritical CO₂. Masoudian [6] also mentioned that the changes in the microstructures of coal could be expected to cause changes in the mechanical properties of solid coal. He used Scanning Electron Microscopy method and found that CO₂ appeared to change the macromolecular structure of the coal. Since the coals are fractured materials with cleats and various pore structures [14], the variance of the microstructures and the change of spatial fracture arrangement can strongly affect the strength of coals. Therefore, all of these former work shows that quantitative description of the variance of microstructures of coal after interaction with supercritical CO₂ is necessary and significant.

During the study of the variance of mechanical properties of coal, many researchers [9,11–13] has mentioned the importance of measurements during in-situ conditions. For the measurement of microstructures, it is extremely hard to do this under in-situ conditions because of experimental technology limits. However, in reality, the injection of CO₂ into coal seam can be cyclic. After each cycle of injection, the desorption of CO₂ will inevitably happen because of CO₂ pressure diffusion and dissipation in the coal seam. Therefore, it is also very meaningful to measure the mechanical property, and microstructure variance before and after displacement of supercritical CO₂. The current experimental methods to measure microstructure of rocks includes mercury injection porosimetry (MIP), low-field nuclear magnetic resonance (LFNMR), scanning electron microscopy (SEM), transmission scanning electron microscope (TEM), small angle scattering (SAS), computed tomography method (CT), and gas adsorption method. All of these methods have their advantages and disadvantages [15]. The MIP is widely used in characterizing the distribution of pore structures, but it is a destructive method. The LFNMR is a recent emerging method in the petrophysical characterization of different rocks, including coals. NMR transverse relaxation (T_2) distributions strongly relate to the coal pore structures [16]. The methods of SEM, TEM, and SAS could show the surface of the microstructures of rocks, but they are limited to the local area. The CT method is a more direct method to restore the whole

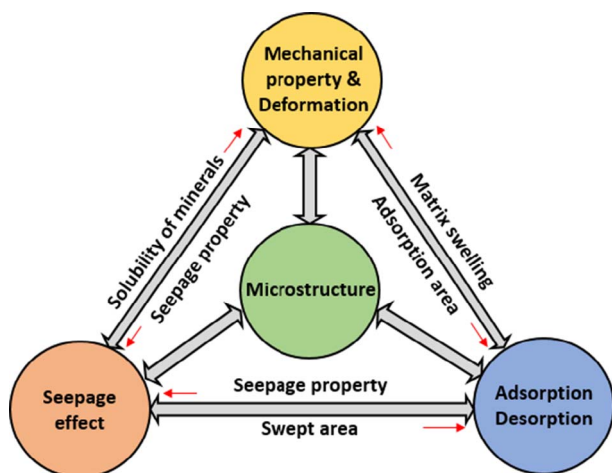


Fig. 1. Multi-factor coupling effect of interaction between supercritical CO₂ and coals.

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