

Perspective

Mechanisms in CO₂-enhanced coalbed methane recovery process

Mohammad Asif¹, Lei Wang²^{*}, Rui Wang², Heng Wang², Randy D. Hazlett¹

¹School of Mining and Geosciences, Nazarbayev University, Nur-Sultan 010000, Kazakhstan

²College of Energy, State Key Laboratory of Oil and Gas Reservoir Geology and Exploitation, Chengdu University of Technology, Chengdu 610059, P. R. China

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Abstract:

Injection of CO₂ and subsequent desorption of CH₄ is considered to be the most efficient enhanced coalbed methane (ECBM) recovery technique to date. Meanwhile, CO₂-ECBM is an excellent option for CO₂ geo-sequestration for an extended period. Despite ongoing research efforts and several field applications of this technology, the mechanisms of the process have yet to be fully understood. The coalbed heterogeneity, the fluid interactions with coal, the CO₂ induced swelling, and the continuous pressure and composition changes require outright insights for optimal application of the technique. Furthermore, intermolecular interactions of CO₂ and CH₄, their competitive adsorption on the dry/wet coal surface, and the dispersion and advection processes play an important role in defining the CO₂-ECBM recovery process. An attempt has been made here to understand the key mechanisms of CO₂-ECBM recovery in coalfields, particularly the adsorption of CO₂ in the supercritical state at the recommended sequestration depth.

1. Introduction

Coalbed methane (CBM) is an important unconventional gas resource under development today (Muther et al., 2022). Nevertheless, 60% to 80% of the adsorbed CBM emits into atmosphere during coal mining (Prabu and Mallick, 2015; Serikov et al., 2022), creating explosion risks, which is traditionally diluted by ventilation. Most of the CBM, e.g., as high as 98%, is adsorbed onto the internal pores with the rest remaining as free gas in cleats of dry coal seams (Naveen et al., 2017; Asif et al., 2019a). The content of gas generally increases with the coal maturity and the depth, i.e., deep high-rank coalbeds contain more gas than shallow coalbeds; however, deep coalbeds are challenging to mine with current technologies (Godec et al., 2014). Unmineable coalbeds, along with basalt formation, saline aquifers, and depleted oil and gas reservoirs, are considered as prospective sites for CO₂ storage to mitigate the rising greenhouse gas level (Bachu, 2000; Naveen et al., 2018; Asif et al., 2022b). CO₂ injection for

enhanced coalbed methane (ECBM), as illustrated in Fig. 1(a), is an effective means to improve the storage economics in coal reservoirs by enhancing CH₄ production and ultimate recovery (Mazzotti et al., 2009). For example, Zheng et al. (2022) reported incremental CH₄ desorption of ~14%-26% from coal powder columns by CO₂ flooding with nuclear magnetic resonance monitoring as compared to natural depressurization; Zhang et al. (2023) tested CO₂-ECBM in a meter-scale coal specimen box and obtained enhancements of ~24%-27% on the basis of conventional CBM recovery. Typically, gases injected for ECBM include N₂ and CO₂. Compared to N₂, CO₂ is preferable owing to its much higher adsorption affinity and higher recovery ability, in addition to future windfalls associated with CO₂ mitigation. Also, CO₂ injection results in better sweep efficiency, whereas N₂ injection causes rapid deformations (Schepers et al., 2010). To date, there still exists a knowledge gap regarding ECBM recovery mechanisms due to complex fluid-solid interacting physics within pores and cleats under reservoir conditions (Asif et al., 2022a).

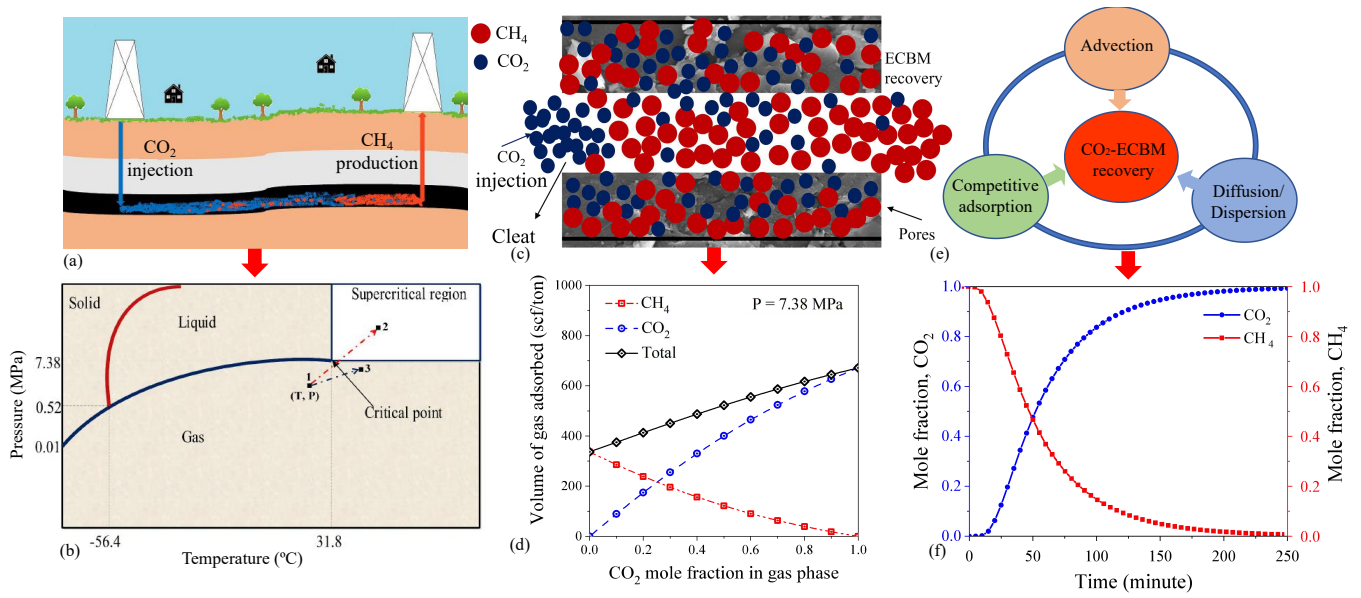


Fig. 1. Key mechanisms in the CO₂-ECBM recovery process (Fig. 1(b) modified after Perera et al. (2011b)).

2. Competitive adsorption in CO₂-CH₄ displacement

To fully understand the CO₂-ECBM recovery process, competitive adsorption in coal should be clarified (Qin et al., 2022). When coal interacts with CH₄ or CO₂, the imbalanced intermolecular forces between the coal atoms on surface enable attraction of other molecules, serving as potential adsorption sites. Competitive adsorption occurs given variations in affinity of different gases towards coal (Ottiger et al., 2008; Asif et al., 2019b). Without decreasing the overall reservoir pressure, the partial pressure of CH₄ can be decreased after CO₂ injection (Shi and Durucan, 2005; Perera et al., 2012), which leads to faster CH₄ desorption from coal surfaces.

2.1 CO₂ phase changes during injection

The recommended minimum depth of coal seams for CO₂ sequestration is around 800 meters, as deeper coal seams provide high enough pressure to keep CO₂ supercritical (Pashin and McIntyre, 2003). There could exist an optimal CO₂ injection pressure in terms of CH₄ recovery and CO₂ storage from the wettability perspective in presence of water (Zheng et al., 2020). Injection modes of constant pressure or stepwise pressurization also affect CBM recovery (Bai et al., 2022). Fig. 1(b) shows the phase diagram of CO₂ with the supercritical pressure and temperature of 7.38 MPa and 31.8 °C. Both temperature and pressure of CO₂ increase with depth from surface conditions (point 1 of Fig. 1(b)) to reservoir conditions, e.g., in deep reservoirs, CO₂ can reach the supercritical state (viz. point 2), whereas in shallow reservoirs, CO₂ would remain as a gas with lower density and storage capacity (viz. point 3). Supercritical CO₂ has a more significant potential to displace CH₄ and acquires higher adsorption capacity than gas state (Perera et al., 2011a).

2.2 Van der Waals equation

The competitive adsorption of CO₂ over CH₄ on coal surface stems from their different physical and chemical properties. The smaller kinetic diameter of CO₂ (0.33 nm) in contrast with CH₄ (0.38 nm) allows CO₂ to enter into all pores where CH₄ molecules reside. In coal matrix, CO₂ or CH₄ molecules adhere to the coal molecules by the weak van der Waals force, their properties can be described by:

$$\left(P + \frac{a}{V^2}\right) \times (V - b) = RT \quad (1)$$

where P is pressure in atm, T is temperature in K, V is gas volume in L/mol and R is the gas constant in L·atm·K⁻¹·mol⁻¹. The van der Waals constant, a , is larger for CO₂ (3.658 L²atm/mol²) than CH₄ (2.30 L²atm/mol²), signifying that CO₂ is preferred over CH₄ for adsorption by molecular attraction. The molar volume of molecules, b , has close values for CO₂ and CH₄.

2.3 Competitive adsorption

The schematic diagram for the CO₂-ECBM displacement and competitive adsorption is shown in Fig. 1(c). The co-adsorption isotherm of CO₂ and CH₄, drawn at the critical pressure and temperature of CO₂, is shown in Fig. 1(d), providing the information about the mole fraction of CO₂ needed for effective ECBM recovery. From Fig. 1(d), it can be deduced that at least 25% of CO₂ in gas phase is required for relatively efficient ECBM recovery, in other words, a 25%/75% CO₂/CH₄ mixture should be expected for the CO₂-ECBM recovery. Moreover, the CO₂ adsorption capacity is 2 to 10 times higher than CH₄ (Gaucher et al., 2011).

2.4 CO₂-ECBM displacement process

As briefly depicted in Fig. 1(e), competitive adsorption, dispersion/diffusion and advection play an important but tan-

gled role together in the displacement process. The dispersion of CO₂ and CH₄ happens in coal matrix during the gas molecule movement between matrix and cleat. In cleat, the advection of CO₂ and CH₄ becomes dominant due to CO₂ injection and movement of mixture (Raouf et al., 2013). The coupled advection and dispersion transport during CO₂-ECBM can be quantified by the well-known advection-dispersion equation, as embodied by the breakthrough curve in Fig. 1(f).

2.5 Leakage risk

Adsorption of CO₂ to coal significantly reduces the chances of backward migration of CO₂ into atmosphere. The leakage of CO₂ may lead to severe accidents similar to the CO₂ gas eruption happened in Lake Nyos in Cameroon in 1986 (Kling et al., 1987). It's desirable to evaluate the fundamental properties of caprock, such as its mineralogy, strength and pore structure, as CO₂ induced reactions, e.g., with calcite, may increase its permeability (Zhang et al., 2019; Xu et al., 2022).

2.6 Matrix swelling and shrinkage

Another key consequence of CO₂ injection into coal is coal matrix swelling, which has profound effects on CO₂ injection, displacement, spreading and sequestration. The matrix swelling or pore space shrinkage is also referred to as a gas sorption-induced strain that reduces the coal cleat width and pore sizes, thereafter decreasing coal porosity and permeability (Reucroft and Patel, 1986; Pekot and Reeves, 2002; Zhou et al., 2011). The sorption-induced strain can contribute up to 60% of the total variation of coal permeability during CBM production (Robertson and Christiansen, 2007; Lu and Connell, 2010). CO₂ influences coal permeability more dramatically than CH₄ on the same concentration basis, e.g., over 90% of permeability reduction could occur after CO₂ injection (Pekot and Reeves, 2002). On the contrary, coal matrix shrinkage happens during N₂ injection, due to its lower adsorption than CH₄. Hence, injection of CO₂-N₂ mixture has been considered as a technical option for ECBM as N₂ suppresses matrix swelling and permeability damage (Zhou et al., 2011).

3. Conclusions and remarks

CO₂-ECBM recovery is a technology that can improve CH₄ recovery as a bridge for energy transition and concurrently sequester CO₂, but knowledge gaps still exist in understanding competitive adsorption and its influential factors. Molecular dynamics is a fast-developing means suitable for visualizing and quantifying the relevant physics in nanoscale processes. Geomechanics, i.e., local changes in porosity and permeability affecting both dispersion and advection in the CO₂-ECBM process. Mineralogy of caprock should be analyzed to have a better strategy to minimize the risks of CO₂ leakage. Matrix swelling and shrinkage could dominate the permeability variation during the injection process, to balance the CO₂ and N₂ percentage becomes necessary for maintaining the injectivity. How injection and production parameters affect the recovery lacks of thorough investigation. On the

whole, more in-depth analyses, experiments, and modeling of CO₂-CH₄-coal interactions are required to more adequately comprehend the promising CO₂-ECBM recovery technology before field implementation.

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Conflict of interest

The authors declare no competing interest.

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