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The mechanical behaviour of coal with respect to CO_2 sequestration in deep coal seams

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Abstract

Carbon dioxide displays a strong affinity for coal due to its propensity to adsorb to the coal surface. The process of CO_2 adsorption on coal causes lowering of surface energy and, it is hypothesised that an associated decrease in surface film confinement results in a decrease in material tensile resistance. Following the results of work carried out on the mechanical influence of CO_2 on brown coal under *in situ* conditions [Viete DR, Ranjith PG. The effect of CO_2 on the geomechanical and permeability behaviour of brown coal: implications for coal seam CO_2 sequestration. Int J Coal Geol 2006;66(3):204–16], a theoretical explanation is proposed for the perceived lack of a weakening effect with the adsorption of CO_2 to coal at significant confining pressures. We propose that at significant hydrostatic stresses, resistance to failure is otherwise provided (by external confinement) and the effects of adsorptive weakening are concealed. Our model predicts that adsorptive weakening, fracturing under *in situ* stresses, and associated permeability increases are not an issue for coal seam CO_2 sequestration for sufficiently deep target seams. Lowering of the elastic modulus of coal upon introduction of CO_2 may proceed by means other than surface energy lowering and could well occur irrespective of the depth of sequestration. The effect of elastic modulus lowering under *in situ* conditions would be beneficial for the long-term retention of sequestered gases.

Keywords: Coal; CO_2 sequestration; Mechanics; Adsorptive weakening; Confinement effects

1. Introduction

In response to predictions of irreversible global warming under current rates of greenhouse gas emission, governments and private institutions alike are considering options to reduce atmospheric emissions. These options include plans to sequester carbon dioxide (CO_2) in large quantities beneath the Earth's surface. The adsorbing nature of CO_2 on coal affords excellent CO_2 retention capacity, a property that has generated interest in the prospect of CO_2 sequestration in unminable coal seams.

Of vital consideration in plans for the large-scale impoundment of CO_2 is the stability of proposed reservoirs. While geotechnical investigation of potential sites would typically involve geological and mechanical characterisation of the reservoir environment, the manner by which CO_2 sequestration modifies the mechanical character of the geological media must also be considered.

The process of adsorption is thought to affect the physical behaviour of solids, and theorisations of the influence of surface-active adsorbents on the mechanical behaviour of solids are well supported by the results of experimental studies [15,6,7]. Of particular relevance to the mechanical influence of CO_2 in the sequestration process are studies concerning the effect of the introduction of CO_2 on the mechanical behaviour of organic substances [8,4,23,1]. While historically, studies have focussed on adsorptive modifications to the material strength, recent investigations have also suggested that the sorption of CO_2 can cause changes in other mechanical properties of coal, specifically, its elastic modulus [22].

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The sequestration environment is very different from those normally considered in coal engineering. Thus the results of conventional coal tests, conducted by investigators interested in coal behaviour under surface or mining conditions, may not be appropriate for direct application to CO_2 sequestration. The role that *in situ* confining pressures play in modifying the influence of CO_2 adsorption on coal seam behaviour is, however, significant. Recently, the results of Viete and Ranjith [22] showed a decrease in both uniaxial compressive strength and elastic modulus with the adsorption of CO_2 on brown coal under atmospheric confinement but showed no strength or elastic modulus decrease for tests carried out under 10 MPa confinement.

This article discusses theory concerning modifications to the influence of CO_2 adsorption on the mechanical behaviour of coal with significant hydrostatic confinement and discusses the origin and consequences for coal seam CO_2 sequestration of changes in the elastic modulus of coal samples exposed to CO_2.

2. Methodology

The results discussed below were drawn from a number of studies concerning the mechanical effects of sorption on solids. The most pertinent results for the current discussion are those of Viete and Ranjith [22], who studied the mechanics of coal seam CO_2 sequestration. They used a uniaxial and triaxial testing approach to investigate the differing mechanical responses of air- and CO_2-saturated brown coal specimens. Overall, four air-saturated and three CO_2-saturated specimens were tested in the uniaxial testing program and tests on four air-saturated and four CO_2-saturated specimens formed the triaxial program. Triaxial runs were carried out at a confining pressure of 10 MPa and internal gas pressure of 2 MPa. Prior to testing, individual specimens were exposed to the appropriate sorbed phase at a pressure of 1.5 MPa for a period of 72 h and, in the case of triaxial tests, internal gas pressures were applied during testing using the gas phase to which the specimen was exposed prior to testing. Uniaxial and triaxial tests used a constant axial strain rate. Viete and Ranjith [22] provide a more detailed description of the testing procedure.

3. Results and discussion

From their results, Viete and Ranjith [22] found a decrease in the compressive strength and elastic modulus of brown coal of about 13% and 26%, respectively, with the introduction of CO_2 for uniaxial tests (Fig. 1a) and no corresponding strength or elastic modulus decrease for the triaxial tests (Fig. 1b). Uncertain of an explanation for the lack of a mechanical response to CO_2 sorption for the specimens tested at larger confining pressures, they suggested that natural mechanical variability in tested specimens might have masked the real effect. Nevertheless, the results of their study provide evidence to suggest that the

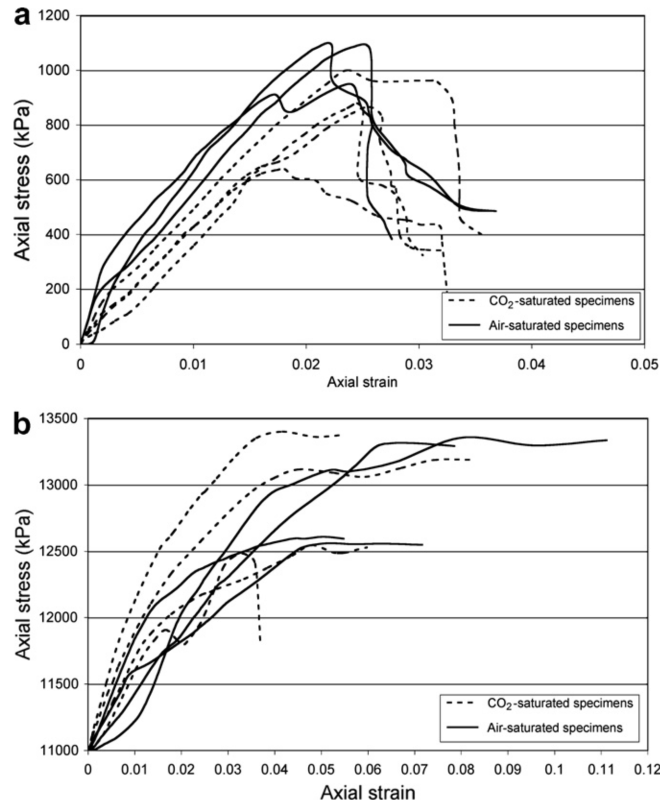

Fig. 1. Stress–strain plots for air- and CO_2-saturated specimens: (a) uniaxial tests, and (b) triaxial tests (from [22]).

adsorption of CO_2 has a negligible effect on the compressive strength and the elastic modulus of coal under significant confining stress.

3.1. The origin of mechanical changes in coal with the introduction of CO_2

The affinity of CO_2 for coal is strongly related to its propensity for adsorption to the coal surface, a process known to influence the mechanical properties of materials (see [19]). However, adsorption is not the only mechanism by which coal can retain CO_2. CO_2 sorption on coal also involves uptake of the sorbent into the coal pore space. Coal is a polymer and the presence of certain functional groups in its polymeric structure allows chemical interaction with solvents (such as CO_2) through electron transfer and a variety of different non-covalent bonds [25,13]. These chemical interactions can cause significant changes to the macromolecular structure of the coal [10,13] and thereby affect its mechanical behaviour [11,25].

An explanation for the apparent lack of strength reduction in the triaxial tests of Viete and Ranjith [22] may be found in adsorption theory. Changes to the coal polymeric structure with the introduction of CO_2 may also play a role in influencing coal strength, though by which mechanisms and to what degree of influence remain unknown. No theoretical explanation for the lack of an elastic modulus decrease under confinement has yet been proposed.

3.2. Changes to coal strength with the introduction of CO_2

3.2.1. Adsorptive weakening

According to Gibbs' adsorption equation [9], a decrease in the surface energy of an adsorbate–adsorbent system will be observed following any change for which the sum of the product of the surface concentration (Γ_i) and the chemical potential ($d\mu_i$) for each adsorbed component increases.

$$d\gamma = -\sum_i (\Gamma_i \, d\mu_i) \tag{1}$$

3.2.2. Surface tension confinement

The Laplace relation gives the compressive stress (σ_s) experienced by a material due to surface tension on a spherical body of radius r [20]

$$\sigma_s = \frac{2\gamma}{r} \tag{2}$$

where γ is the surface energy.

Yates [26] and Dolino et al. [5] discuss Eq. (2) with respect to the shrink–swell behaviour of solids with adsorption, a phenomenon first noted for CO_2 adsorption on charcoal by Meehan [12].

Though Eq. (2) is specific to the case of a spherical body, it is proposed that the surface tension-related compressive stress, it describes, will be observed for any adsorbate in the presence of a surface-active adsorbent, its degree of influence dictated by the geometry the adsorptive film takes. Scherer [18] provides expressions for compressive stress modification with changes in surface energy for more complex geometries.

We advocate the notion that changes to the surface energy of a solid-adsorbent system affects the degree of confinement that surface tension exerts and therefore the stress that must be applied to cause the solid to fail under tension. The term 'tensile strength', as it will be henceforth used, should not be viewed as the strength of the material in isolation, but rather the applied stress that will cause tensile failure of a solid under the prevailing sorption environment.

3.2.3. Confining stress modifications to adsorptive weakening

If tensile failure is to occur in a material, tensile stresses must locally exceed the confinement-induced compressive stresses that act to resist tensile failure. Confining stresses may have extrinsic origins (e.g. hydrostatic stresses below the earth's surface) or may be intrinsic to the material-sorbent system (e.g. confinement by surface tension in adsorbed films). Where pressures related to external confinement (σ_e) exceed those inherent to the material-sorbent system (σ_i), modifications that lower σ_i will not decrease the applied pressure required to cause failure in that material, as resistance to tensile failure is otherwise provided by externally derived compressive stresses. On the other hand, for situations where σ_i is greater than σ_e, material changes that produce a decrease in σ_i, such as surface energy lowering according to Eqs. (1) and (2), also cause a corresponding decrease in the tensile strength of the material (Fig. 2).

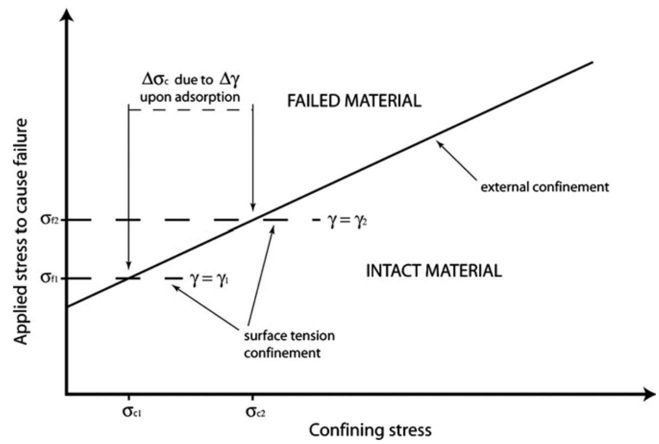

Fig. 2. Arbitrary plot showing applied stress to cause failure vs. confining stress. With adsorbent exchange and a change in surface energy from γ_2 to γ_1, there is a decrease in the external confining stress below which surface tension provides the confinement (from σ_{c2} to σ_{c1}) and in the applied stress required to cause failure (from σ_{f2} to σ_{f1}) under modest confinement. For an external confining stress larger than σ_{c2}, adsorbent exchange will have zero influence on the applied stress required to cause failure.

For values of hydrostatic pressure smaller than the confining stress imparted by surface tension under lowered surface energy conditions (post-adsorbent exchange), the full effect of CO_2 adsorptive weakening on coal is expected, while for hydrostatic pressures greater than surface tension-related confining stresses at initial adsorbent concentrations (prior to CO_2 saturation), no weakening is predicted. We suggest that, in the latter case, any role that surface tension plays in strengthening the material against tensile failure is nullified by the larger confining stresses sourced from the weight of the overlying material; external confinement conceals the apparent weakening effect.

Quantification of the proposed confining stresses at which CO_2 weakening on coal becomes negligible requires an intimate knowledge of CO_2 adsorption kinetics for a given coal seam and is highly dependent on coal pore-space geometry, rank and moisture content and the composition and abundance of initial coal seam gases with respect to the sequestered gas. Despite these dependencies, Ravikovitch and Neimar [16] have suggested that adsorption-induced stresses in carbon-based materials can be to the order of megapascals. The results of Viete and Ranjith [22] indicate that the strength decrease due to CO_2 adsorption on brown coal from the Latrobe Valley of southeast Australia is negligible at confining pressures of 10 MPa, equivalent to a coal seam depth of about 400–500 m.

3.3. Changes to the elastic modulus of coal with the introduction of CO_2

In addition to their influence on material strength, sorption-related modifications can also cause changes in the elastic properties of materials. When certain materials are subjected to long-term water sorption, a decrease in the elastic modulus has been observed. Obataya et al. [14], Badens et al. [2] and Rysiakiewicz-Pasek et al. [17] noted a decrease in the elastic modulus of spruce timber, set plaster and porous glass, respectively, with an increase in the moisture content. Wang and Kramer [24] noted a significant decrease in the elastic modulus of polystyrene with the introduction of high pressure CO_2 gas. Viete and Ranjith [22] observed a decrease in the elasticity of brown coal of 26% with the introduction of CO_2 as a sorptive phase in the case of uniaxial tests (Fig. 1a) but no elastic modulus decrease for their triaxial tests (Fig. 1b).

Wang and Kramer [24] suggested that the decrease in elastic modulus of polystyrene they observed with the introduction of pressurised CO_2 was related to plasticisation of the sorbate, a process brought about by CO_2-triggered changes to the polymeric structure of polystyrene. Smith and Moll [21] reported a similar plasticisation effect of CO_2 on polycarbonate, polyester carbonate and polystyrene. Larsen [11] and White et al. [25] have proposed that coal, being a polymer, will also experience changes in its polymeric structure and associated plasticisation with absorption of CO_2. What is now of interest is whether confining stress plays any role in influencing this plasticisation effect. It is clear that weakening and plasticisation of coal

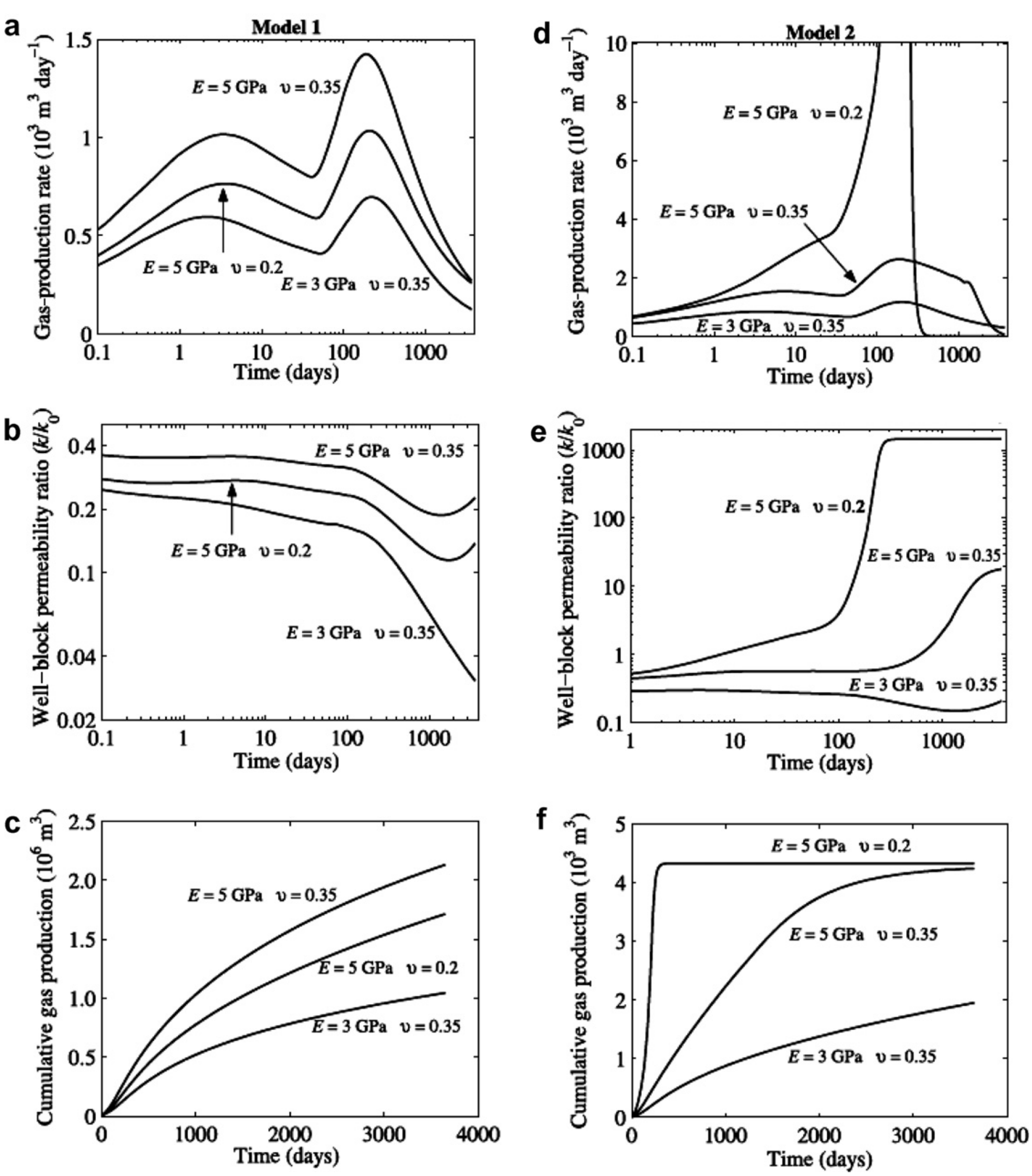

Fig. 3. The influence of mechanical properties of a coal seam (E = elastic modulus, υ = Poisson's ratio) on: (a) and (d) coal-bed gas production rates; (b) and (e) well-block permeability; and (c) and (f) cumulative gas production (from [3]). Plots (a)–(c) are specific to Model 1 and plots (d)–(f) are specific to Model 2 of Cui and Bustin [3] which, respectively, account for and fail to account for sorption-induced strain.

with the addition of CO_2 do not proceed (at least not exclusively) by the same mechanism and, in spite of the results of Viete and Ranjith [22], there is no theoretical evidence to suggest that the plasticisation effect is suppressed by physical confinement.

Adsorptive weakening does not appear to be of concern for CO_2 sequestration in coal seams at sufficient depth; however, the effects of coal elastic modulus lowering by CO_2 introduction may be influential. Thus, the consequences of elastic modulus lowering for coal seam CO_2 sequestration merits consideration. Cui and Bustin [3] used a numerical modelling approach to study the effect of coal properties on rates of coal-bed methane production. They provide data to suggest that long-term coal seam permeability will decrease with a lowering of the elastic modulus of the coal seam (Fig. 3) and interpret this as being due to the diminished ability of the host material to retain coherence with the removal of pore-space gas. In terms of permeability reduction, a larger response can be expected for a given increase in effective stress in the presence of CO_2 than for the same effective stress increase in the presence of a pore-space gas such as methane. The coal would thus show a greater tendency toward regulation of pore-space gas flow where CO_2 represents a larger proportion of the sorbed phase. This would suggest that a propensity for coal plasticisation with the introduction of CO_2 is beneficial for the CO_2 sequestration process in coal.

4. Conclusion

Adsorptive surface energy lowering and associated weakening by the introduction of CO_2 to coal can present significant problems under surface or near-surface conditions. At significant pressures of confinement these effects appear to be concealed. We propose that CO_2 adsorption on coal causes a change in material confinement related to surface tension. Accordingly, increases in coal seam permeability due to adsorptive weakening and fracturing under *in situ* stresses are not likely to present a risk where coal seam CO_2 sequestration is carried out at sufficient depths. Coal elastic modulus lowering by CO_2 sorption could be independent of environmental pressures and must, at this stage, be considered an issue of importance in CO_2 sequestration in deep coal seams. With a decrease in the elastic modulus of coal, a coal seam is inclined to further regulate pore-space flow by greater permeability lowering in response to gas migration and associated localised increases in effective stress. The validity of the notions promoted in this paper await verification through further laboratory testing and field trials.

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