Constitutive equations for coal containing gas considering gas adsorption

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Abstract

Coupled gas flow and solid deformation processes in porous media have received considerable attention because of its importance in CBM exploitation and gas outbursts during coal mining. Coalbed methane is mainly adsorbed on coal. The dependence of gas adsorption on gas pressure has significant effect on the mechanical properties of coal, however, the effect was ignored in most of the previous modeling studies. Based on Truesdell’s postulate of the theory of mixtures, a new set of constitutive equations for the saturated mixtures of gas-filled coal which is composed of coal matrix, adsorbed gas and free gas is developed. Adsorbed gas pressure and the volume fraction of each component of the saturated mixtures are also presented. The proposed constitutive equations show that mechanical response of coal is determined by stress condition, mechanical properties of solid phase, gas pressure, gas adsorption, pore structure and its evolution. Without considering the adsorption effects and pore evolution, the effect of gas pressure in coal seam would be underestimated.

Keywords: coal containing gas; constitutive equations; adsorption; the theory of mixtures

1. Introduction

Coupled gas flow and solid deformation processes in porous media have received considerable attention because of its importance in CBM exploitation and gas outbursts during coal mining. Coalbed methane is mainly adsorbed on coal. Adsorption and desorption of gas can cause volumetric deformation of coal\cite{1, 2}. A series of measurements of adsorption-induced coal swelling show that the volumetric strain and pressure can be described by using a Langmuir-like equation, and that the volumetric strain is approximately linearly proportional to the amount of gas adsorbed\cite{3, 4}. Swelling of cylindrical coal samples subjected to sorption–desorption cycle by employing strain gauges and acoustic transducers indicate that coals possess anisotropic property in swelling and the degree of swelling rate is larger in the direction perpendicular to the bedding plane than that parallel to the bedding plane\cite{5}. This property and the anisotropic structure of coal are attributed to the stress conditions under which the coal was formed\cite{6, 7}.

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In previous works, coal strength and deformation modulus have been found to change with adsorption of carbon dioxide (CO₂), methane (CH₄) and other gases⁸–¹³, showing that adsorption modifies the physical structure of the coal matrix and may even extract some of the polycyclic aromatic hydrocarbons and mobilize them in the coal seam¹⁴. The modification in physical structure is associated with a relaxation and rearrangement of the macromolecular structure of the coal. The molecular rearrangement of coal caused by molecules dissolving in the coal can be explained by classical polymer chemistry. Coals are glassy, strained, cross-linked macromolecular systems that are not at the lowest energy state⁶. The brittleness of coals is due to their glassy structure, which has intra-molecular interactions greater than the available thermal energy, and the molecules have limited freedom to move except for some small-scale vibrations and rotations. When the coal interacts with a solvent, the free volume of the polymeric system increases and glass transition temperature drops (the temperature at which the glass becomes a rubber).

Therefore, adsorption of gas may play an important role in the stress-strain relationship, the strength of coal, the speed at which a coal can adsorb additional gas. Such changes impact commercial production of coalbed methane, potential degasification of future mining areas by drilling horizontal and vertical degasification wells, enhanced methane recovery using CO₂ injection, and concurrent sequestration of CO₂¹⁵. Investigation on adsorption effect can also provide an understanding of the mechanisms leading to gas outbursts in underground coal mines.

Recently, Jiang et al.¹⁶ proposed a new set of constitutive equations for saturated mixtures of coal containing methane based on the theory of mixtures, which shows that mechanical properties of coal containing methane are determined by properties of coal constituents, pore distribution and adsorption / desorption characteristics of gas. As the continuation and development of the above research, this study presents a stress-strain relation to describe mechanical characters under triaxial stresses. And the volume fraction of each constituent of the saturated mixtures is derived. The aim of this study is to investigate the deformation behavior of a confined and stressed coal under gas adsorption/desorption.

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2. The constitutive equations

2.1. Basic hypothesis

According to previous research achievements, considering the couple effect between gas and coal, the basic hypothesis of the constitutive model is established based on Truesdell’s postulate of the theory of mixtures.

Firstly, coal containing gas, which is composed of solid-phase components, adsorbed gas and free gas, is viewed as a saturated mixture. A mixture is defined to be saturated if:

\[ \sum_i \phi_i = 1 \]  

(1)

For any material volume \( dV \) of the mixture, a portion of that volume \( dV_i \) is occupied only by the \( i \) th constituent. The volume fraction \( \phi_i \) is given by

\[ \phi_i = \frac{dV_i}{dV} \]  

(2)

Therefore we will pay particular attention to each phase in coal containing gas, which consists of a porous solid, \( i = s \), a gas, \( i = g \), and adsorbed gas, \( i = a \).

As a result of the mass between gas phase and solid phase cannot be transformed into each other, in a control room, solid phase follows mass conservation law of a closed system. And the natures of solid phase are homogeneous and isotropic, which determined by those of its components. Secondly, it is assumed that the process of adsorption and desorption is instantaneous and isothermal.

Thirdly each component of coal containing gas has the same temperature. And gas adsorption amount of coal samples from same sampling spots keeps invariant under the condition of same gas pressure and components.

2.2. The constitutive equations of coal containing gas

According to the conservation equation of mixture theory, the general stress tensor of coal containing gas can be expressed as:

\[ \sigma = \phi_s \sigma_s - \phi_g \bar{p} I - \phi_a \hat{p} \tilde{a} I - \dot{\phi}_g (\sigma_g + p I) - \dot{\phi}_a (\sigma_a + p_a I) \]  

(3)

where \( \sigma_s, \sigma_g, \sigma_a \) are the stress tensor of mixture and the solid phase. \( p \) and \( p_a \) mean the pore pressure and adsorbed gas pressure, respectively. Notice that the subscripts \( s, g, a \) represent solid, gas and adsorbed gas.

The above constitutive equations embody dynamic balance and transform between adsorbed gas and free gas, in which volume fractions are important. Furthermore volume fraction of adsorbed phase is difficult to confirm. It shows that the density of the adsorbate is constant in the gas phase and constant (zero) within the solid, and that the density of adsorbed phase change with the location of its boundaries. The properties of adsorbed molecules are different from that of gas-phase molecules, but ambiguity arises for distinguishing the two forms. Different from materials containing non-adsorptive gas, coal will swell as a result of gas adsorption. So volumetric swelling is viewed as existence space of adsorbed phase. According to basic hypothesis of coal containing gas, volume fraction of each constituent can be calculated as

\[ \phi_s = \frac{1 - n^*}{1 + 3 \varepsilon_a}, \phi_g = \frac{n^*}{1 + 3 \varepsilon_a}, \phi_a = \frac{3 \varepsilon_a}{1 + 3 \varepsilon_a} \]  

(4–6)

where \( n^* \) is porosity of mixture containing non-adsorptive gas under setting stress state and \( \varepsilon_a \) is swelling caused by adsorption of gas alone.

Substituting Eq. (4–6) to Eq. (3) yields
Assuming the stress-strain relations of solid phase obey generalized Hooke’s Law, we obtain from Eq(7)

\[
\Delta \varepsilon - \Delta \varepsilon^* = \frac{(1 + 3\varepsilon_a)^2}{(1 - n^*)^2} E_s \left[ \frac{n^*}{1 + 3\varepsilon_a} \Delta p + \frac{3\varepsilon_a}{1 + 3\varepsilon_a} \Delta p_a + \left( \frac{n^*}{1 + 3\varepsilon_a} - \frac{n^*}{1 + 3\varepsilon_a} \right)^2 \right] \]

\[
\Delta \varepsilon - \Delta \varepsilon^* = \frac{(1 + 3\varepsilon_a)^2}{(1 - n^*)^2} E_s \left[ \frac{n^*}{1 + 3\varepsilon_a} \Delta p + \frac{3\varepsilon_a}{1 + 3\varepsilon_a} \Delta p_a + \left( \frac{n^*}{1 + 3\varepsilon_a} - \frac{n^*}{1 + 3\varepsilon_a} \right)^2 \right] \]

where \( \Delta \sigma_r, \Delta \sigma_z \) are the axial and radial stress increment of mixtures, \( \Delta \varepsilon_r, \Delta \varepsilon_z, \Delta \varepsilon_v \) are the axial, radial and volumetric strain increment of mixtures. And the parameter \( \Delta \varepsilon^* \) and \( \Delta n^* \) obey

\[
\Delta \varepsilon^* = \frac{1 + 3\varepsilon_a}{1 - n^*} \Delta \varepsilon_a, \quad \Delta n^* = \frac{1 - n^*}{1 + 3\varepsilon_a} \Delta \varepsilon_r - \Delta \varepsilon_v - 3\Delta \varepsilon_a
\]

Eq. (8) and Eq. (9) show that mechanical response of coal is determined by stress condition, mechanical properties of solid phase, gas pressure, gas adsorption, pore structure and its evolution. Changing axial stress alone and using Eq. (8) and Eq. (9) lead to

\[
\Delta \varepsilon_r = \frac{(2\varepsilon_r - 1)\Delta \sigma_r}{2 \left( \frac{1 - n^*}{1 + 3\varepsilon_a} \right)^2 E_s} \left( \sigma_r + 2\sigma_z + \frac{3}{1 + 3\varepsilon_a} p_a + 9\varepsilon_a p_a \right)
\]

\[
\Delta \varepsilon_r = \frac{(1 + 3\varepsilon_a)^2}{(1 - n^*)^2} E_s \left[ \Delta \sigma_r + \frac{1 - 2\varepsilon_r}{(1 + 3\varepsilon_a)^2} \Delta \sigma_r + \frac{\Delta \varepsilon_r}{1 + 3\varepsilon_a} \left( \frac{3}{1 + 3\varepsilon_a} p_a \right) \right]
\]

\[
\Delta \varepsilon_r = \frac{(1 + 3\varepsilon_a)^2}{(1 - n^*)^2} E_s \left[ -\varepsilon_r \Delta \sigma_r + \frac{1 - 2\varepsilon_r}{(1 + 3\varepsilon_a)^2} \Delta \sigma_r + \frac{\Delta \varepsilon_r}{1 + 3\varepsilon_a} \left( \frac{3}{1 + 3\varepsilon_a} p_a \right) \right]
\]

Nevertheless, the deformations of the solid phase and adsorbed phase have been neglected in the above equations.
3. Results and Discussion

Authors obtained CO$_2$ and N$_2$ isothermal adsorption curves and the porosity of the samples from Yangquhe mines. The adsorption measurement is up to 10.0 MPa. Meanwhile, author measured the stress-strain curve of the samples containing CO$_2$ and N$_2$ under triaxial compression. The confined pressure is 5MPa, the pore pressure is 2MPa and loading rate is 0.0035MPa/s. Fig.1 shows the observed stress-strain curves of standard coal samples containing CO$_2$ and N$_2$ with respect to the difference of axial stress. The experiment parameters are given in Table 1.

Table 1. Summary of parameters for coal samples

<table>
<thead>
<tr>
<th>Gas type</th>
<th>$L$ (cm$^3$/g)</th>
<th>$B$ (1/MPa)</th>
<th>$n$ (%)</th>
<th>$E$ (MPa)</th>
<th>$\nu$</th>
</tr>
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<tbody>
<tr>
<td>N$_2$</td>
<td>18.81</td>
<td>0.365</td>
<td>6.76</td>
<td>3800</td>
<td>0.29</td>
</tr>
<tr>
<td>CO$_2$</td>
<td>54.36</td>
<td>0.353</td>
<td>6.76</td>
<td>3507</td>
<td>0.35</td>
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</tbody>
</table>

The other parameters needed for Eq. (12), Eq. (13) and Eq. (14) include the adsorbed gas pressure, elastic modulus Poisson’s ratio of solid phase and adsorption-induced swelling, which were estimated and calculated from the experiment data. The relationships between $E_s$ and $E$, as well as $\nu_s$ and $\nu$ are given as

$$E = \frac{E_s \rho}{(3\rho - 2\rho)}, \quad \nu = \nu_s + \frac{3(1-\nu_s^2)(1-5\nu_s)n}{2(7-5\nu_s)} \quad (15-16)$$

The $E_s$ is assumed to be 4200 MPa and $\nu_s$ to be 0.35, which are reasonable for coals. The adsorbed gas pressure can refer to

$$p_a V_m = RT \left( 1 + \frac{B}{V_m} + \frac{C}{V_m^2} \right) \quad (17)$$

where $V_m$ is molar volume of adsorbed gas; $B$ and $C$ are the second and third Virial coefficients, respectively, which are assumed to be -40.91 cm$^3$/mol and 2320 cm$^6$/mol$^2$.

Assuming isotropic elastic behavior and that the change of the elastic energy equals to the change of surface energy, Pan obtained the swelling strain for unit mass of absorbent and given as

$$\varepsilon_s = RT \ln \left( 1 + b p_a \right) \frac{\rho_s}{E_s} f(x,\nu_s) - \frac{3f(x,\nu_s)}{(1-n)E_s} \int_0^{\rho_s} \varepsilon_s dp \quad (18)$$

where $E_s$, $\rho_s$, and $\nu_s$ are the elastic modulus, density, and Poisson’s ratio of solid phase, respectively; $R$ and $T$ the gas constant and temperature; $L$ and $b$ the Langmuir constants; and $f(x,\nu_s)$ is a parameter related with porosity and Poisson’s ratio of solid phase, which can be written as

$$f(x,\nu) = \frac{\left[2(1-\nu_s) - (1+\nu_s)c x \right] \left[3-5\nu_s - 4(1-2\nu_s)c x \right]}{(3-5\nu_s)(2-3c x)} \quad (19)$$

where $c = 8\sqrt{2}/3\pi = 1.200$ and $x = a/l$. From Pan’s model, porosity can be calculated as

$$n = 1 - 3\pi x^2 (1-c x) \quad (20)$$
Fig. 1. The stress-strain curves of triaxial compression tests  
Fig. 2. The stress-strain curves from derived constitutive equations

Fig. 2 presents the calculated stress-strain curves with respect to the difference of axial stress. The numerical results indicate the swelling of coal samples increases and the deformation modulus decreases with the amount of adsorbed gas. The calculated results are in agreement with the experimental data, especially for axial strains.

4. Conclusions

1) Based on the theory of mixtures, the constitutive relation of coal containing gas is derived, in which the effects of adsorption are introduced and the parameters have clear physical meaning. From the theoretical equations, the strain of coal containing gas is determined by not only the stress state and mechanical properties of solid phase, but also the volumetric strain due to adsorption, the porosity and its evolution.

2) The constitutive relation can be used to describe the coal deformation behaviors related to adsorption. The numerical results dedicate the deformation modulus of coal sample decreases with the amount of adsorbed gas. Without considering the adsorption effects and pore evolution, the effect of gas pressure in coal seam would be underestimated.

Acknowledgements

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