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Experimental Investigation on Strain Changes during CO_2 Adsorption of Raw Coal Sample: Temperature and Effective Stress

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Abstract: Through laboratory simulation experiments, this paper studies the influence of different temperature and stress conditions on strain changes of raw coal samples induced by the CO_2 adsorption with tri-axial creep-seepage and adsorption-desorption experimental system. Comparing and analyzing the experimental results, the study shows that: (1) within a certain time, the axial and radial strain of the raw coal sample induced by CO_2 adsorption both show a growing trend as the adsorption time increases and the strain of the raw coal sample for CO_2 adsorption is obvious anisotropy; (2) at the same point in time, the greater the axial effective stress, the smaller the axial strain increasing rate of the loaded coal sample during CO_2 adsorption process and the smaller the value of axial deformation; (3) during the adsorption process, the volume strain of raw coal sample decreases with the increasing of temperature, namely, the adsorption capacity of raw coal sample decreases with the increasing of temperature.

Keywords: raw coal sample; CO_2 adsorption; effective stress; temperature effect; volume strain

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1. Introduction

With the increasing severity of the greenhouse effect, as the most important climate-related greenhouse gas in the Earth's atmosphere [1], the large amount of CO_2 released by human production and living activities and the consumption of traditional fossil fuels has attracted more and more attention. Meantime, as a new green technology, the utilize of CO_2 has made great progress in unconventional natural gas exploitation, especially in depleted gas reservoirs [2] and coalbed methane [3,4]. A large number of CO_2-ECBM (CO_2 enhanced coal bed methane recovery) pilot experiments and previous studies have been carried out all over the world. Ranathunga, A.S experimentally studied the CO_2-ECBM potential of low-rank coal under different CO_2 injection conditions at mesoscale [5]. Wang [6] studied the adsorption and diffusion behaviors of CH_4 and CO_2 in coal seams by molecular simulation. Prusty, B.K [7] studied and evaluated the effect of CO_2 injection on coalbed methane production and CO_2 storage potential by using coal samples from three different coal seams and considered that CO_2 injection has different enhanced gas recovery patterns for different coal seams. Besides, some numerical simulation studies [8,9] and others [10] all showed that it is one of the important ways to reduce carbon emissions and increase coalbed methane production by injecting CO_2 (Gaseous carbon dioxide), LCO_2 (Liquid carbon dioxide) [11] and SCO_2 (Supercritical carbon dioxide) into coal seams by feasible means to promote the expansion of coal reservoir pores and fissures, change the stress state of coal reservoirs [12], displace CH_4 [13,14] and realize the permanent storage of CO_2 [15]. Consequently, it is of great significance to deal with the global greenhouse effect, improve the production of coalbed methane and even reduce the coalbed methane disasters.

Although the way of CO_2 injection into coal reservoir can greatly increase the production of coalbed methane by replacing CH_4, the adsorption deformation effect caused by CO_2 adsorption on coal body will cause the obvious changes of gas migration path in coal reservoir, which results in difficulties in coalbed methane production, the sharp decline in production and other questions. Therefore, it is the key and basis to study the influencing factors and changing laws of adsorption strain during the project of enhancing coal bed methane recovery by injecting CO_2 and realizing CO_2 geological sequestration. In response to this problem, many scholars have conducted some experimental studies on the influence of CO_2 injection into coal reservoirs on permeability mainly from temperature, stress and injection pressure. Wang [16] studied the effects of different gas pressures and temperatures on the permeability and steady-state time of anthracite coal using a self-developed steady-state permeability test system for coal and rock. It is believed that temperature influences the permeability of coal by changing the thermodynamic behavior, gas molecular dynamic movement and adsorption-desorption process. Through setting temperature and pressure conditions, Liu [17] carried out experimental research on cuboid raw coal samples with the size of 70 × 70 × 300 mm, which proved that CO_2-ECBM technology was feasible in theory and verified that CO_2 injection had a superior effect in enhancing coalbed methane recovery. Zhang [12] conducted a series of CO_2 core displacement tests on high-grade bituminous coal cores with an injection pressure of 6–10 MPa. It is concluded that the volumetric strain caused by CO_2 adsorption is approximately proportional to the storage capacity of CO_2 in coal samples and it increases with an increase in CO_2 pressure. These scholars have emphasized the important role of stress, temperature and CO_2 pressure during CO_2 injection into coal reservoirs.

In the project of CO_2-ECBM and CO_2 geological sequestration, the phase state, the pressure and the temperature of CO_2 injected into coal reservoirs have a great influence on the injection effect. At the same time, with the change of geological conditions, especially the temperature and stress conditions of coal reservoir, the state of CO_2 injected into coal reservoirs changes accordingly and then which in turn changes the CO_2 adsorption-desorption strain response and the permeability [18]. Therefore, this paper mainly studies the deformation response law of coal samples in the process of CO_2 adsorption by changing the CO_2 temperature and pressure and the axial effective stress conditions of experimental coal samples and it experimentally reveals the influence of temperature and stress conditions on strain changes in the process of CO_2 adsorption by raw coal sample, which has certain reference significance for CO_2-ECBM and CO_2 geological sequestration engineering.

2. Samples and Experimental Process

2.1. Samples

The raw coal samples used in this paper are taken from the Zhujiao coal mine in Anyang City, Henan Province in China (Figure 1). Firstly, the large coal samples (a) were taken from the driving roadway 750 m underground. Secondly, the large coal samples were sealed with insulating ester and transported back to the laboratory. Finally, the cylindrical raw coal samples (b), (c) with a height of 100 mm and a diameter of 50 mm were prepared by using a special core drilling machine. In order to avoid the influence of physical and mechanical properties [4,19,20], anisotropy [20–22] and other conditions of raw coal samples on coal adsorption strain, the cylindrical raw coal samples of this paper are drilled along the vertical bedding direction. Some of the coal samples were put into the drying oven for 48 h (60 °C) and then sealed for storage. Some prepared coal samples (c) were taken for testing and the test results show that the proximate analysis and basic data of raw coal samples in Tables 1 and 2 and the average values of their moisture content, volatile content, ash content and fixed carbon are 0.55%, 24.33%, 12.17% and 63.20%, respectively.

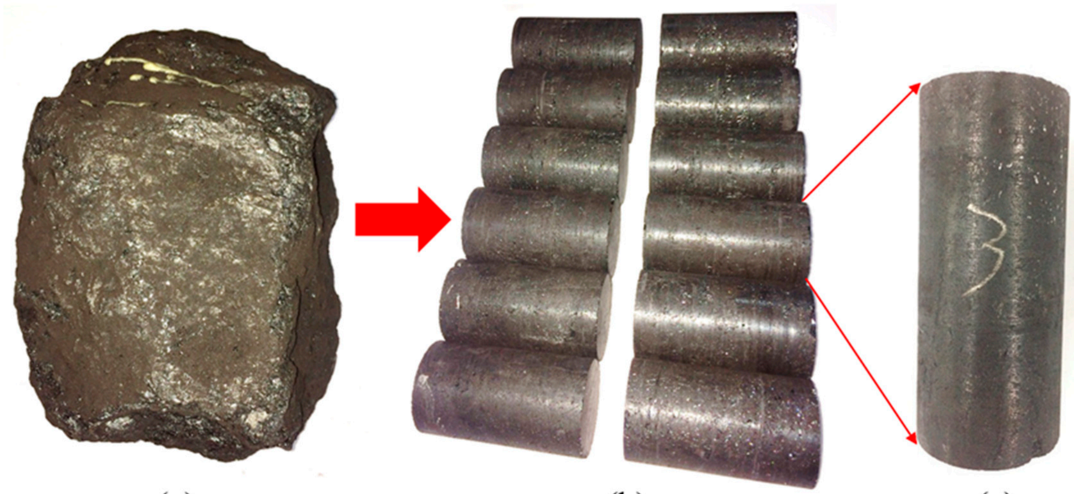

Figure 1. Raw coal sample for the experiment: (**a**) large pieces of raw coal from coal mine; (**b**) cylindrical coal samples; (**c**) the raw coal sample used in the experiment.

Table 1. Proximate analysis results of dried raw coal samples.

Sample Label	Mad/%	Vdaf/%	Ad/%	Fcd/%
#1	0.55	24	12.12	63.33
#2	0.55	24.08	12.04	63.33
#3	0.54	24.46	12.25	62.75
#4	0.56	23.76	12.29	63.39

Mad, air-dried moisture; Ad, dry ash; Fcd, dry fixed carbon; Vdaf, dry ash free.

Table 2. The average value of basic data of dried raw coal sample.

Sample Label	Weight/g	Length/mm	Diameter/mm	Volume/cm^3	Density/g/cm^3	Permeability/md
#	247.80	96.70	49.62	186.90	1.33	0.0174

2.2. Experimental Equipment and Procedures

This paper mainly studied CO_2 adsorption strain variation of the loaded coal sample under temperature and axial stress through laboratory simulation experiments. The equipment used in the experimental research comes from State Key Laboratory Cultivation Base for Gas Geology and Gas Control of Henan Polytechnic University, mainly including the tri-axial creep-seepage and adsorption-desorption experimental systems of coal and rock (Figures 2 and 3), Figure 2 is the schematic diagram of the tri-axial creep-seepage and adsorption-desorption experimental systems of coal and rock, Figure 3 is the physical diagram of the alcohol circulating refrigeration apparatus in the coal sample chamber and the tri-axial creep-seepage and adsorption-desorption experimental systems of coal and rock. Based on the existing experimental equipment, the temperature adjustment device (4, 14) and the gas injection booster pump (Z) are added to achieve the CO_2 temperature and CO_2 gas pressure conditions required for the experiment. The data required in the experiment can be monitored and collected in real-time by pressure sensors (P) and strain sensors (9, 10, 11).

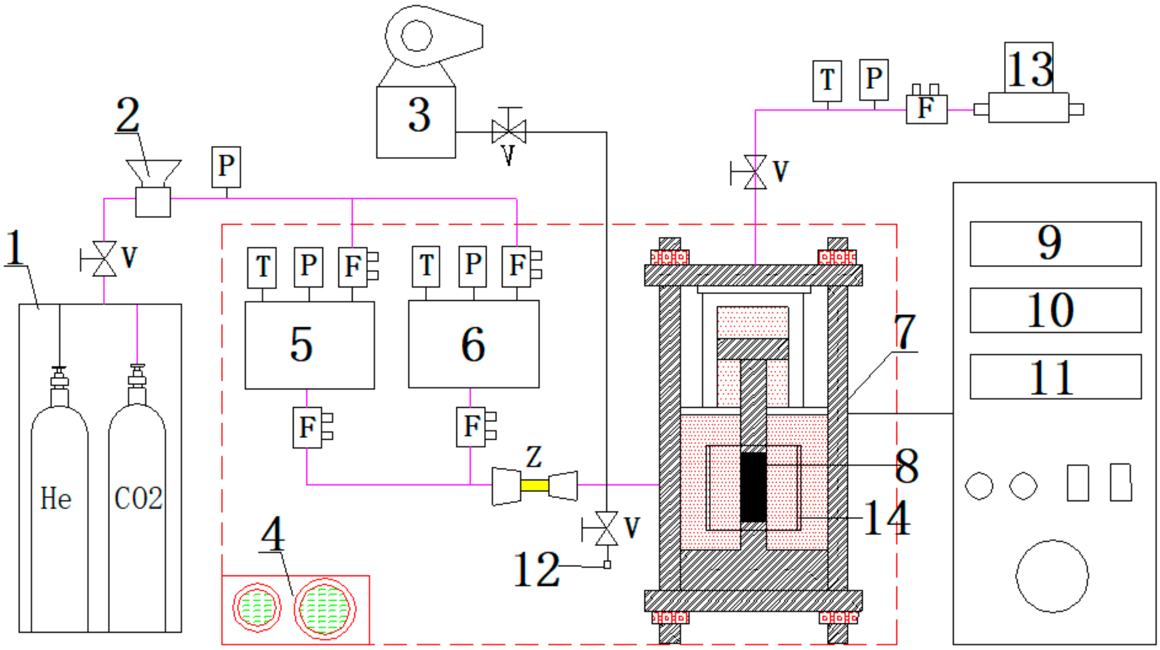

Figure 2. The schematic diagram of the tri-axial creep-seepage and adsorption-desorption experimental systems of coal and rock.

Figure 3. The physical diagram of the alcohol circulating refrigeration apparatus in the coal sample chamber and the tri-axial creep-seepage and adsorption-desorption experimental systems of coal and rock: 1-high-pressure gas tank group; 2-pressure regulating valve; 3-vacuum pumping device; 4-temperature control device, mainly including ethanol circulating refrigeration device and electric heating device; 5–reference tank group, system transfer gas supply device; 6-standard tank group, system calibration gas supply device (He); 7-coal sample chamber, sample loading and fixture device; 8-sample, this time is coal sample; 9-axial stress loading servo control device; 10-radial stress loading servo control device; 11-pore pressure servo control device; 12-system exhaust device; 13-gas flow meter group; 14-the alcohol circulating refrigeration apparatus; F-air valve; P-pressure sensor; T-Temperature sensor; Z-gas booster device.

As shown in Figure 2, the main experimental process of this paper is as follows: CO_2 gas from the high-pressure gas tank group (1) entered the gas source through the pressure regulating valve (2), then entered the coal sample (8) in the coal sample chamber (7) through the gas booster device (z) and finally discharged through the gas flow meter group (13). During the experiment, the temperature control device (4) and the alcohol circulating refrigeration apparatus (14) were responsible for adjusting the temperature of the main experimental devices and pipes and the axial stress loading servo control device (9) and the radial stress loading servo control device (10) were responsible for loading the coal samples.

Before the beginning of experiments, it was necessary to check and confirm that the airtightness of the entire experimental system was intact and then the dead space (pipeline fixed space and tank group fixed space) of the experimental system was measured and recorded by using the gas in the standard tank group. And then the prepared experimental coal sample was put into the coal sample room and sealed and fixed by liquid silicone rubber and heat shrinkable tube to prevent air leakage and the hydraulic fluid in the oil cavity from polluting the coal sample. After solidifying of the liquid silicone rubber on the side surface of the coal sample, the axial deformation gauge and radial deformation gauge were installed on the coal sample, At the same time, it was connected that the gas inlet and outlet pipelines of the coal sample and then the predetermined axial load and radial load were loaded. Secondly, the inlet and outlet valves of the system were closed and then the whole experimental device was vacuumized for 12 h to reduce the influence of other gases on the experiment. Simultaneously, the airtightness and vacuum effect of the experimental device could be judged by observing the reading of the vacuum gauge. Finally, the coal sample chamber reached the predetermined temperature and CO_2 gas pressure to meet the experimental conditions required for the research by adjusting the temperature control device and the gas injection booster pump. Then, CO_2 of the stated pressure was continuously injected into the coal sample chamber for adsorption and the adsorption time was 8 h. During the entire adsorption process, the axial and radial strain changes of the experimental coal sample were synchronically monitored and recorded. After the adsorption, the system inlet valve was closed and the outlet valve was opened for desorption of the raw coal sample. the experimental research would be completed by repeating the above operations with changing the predeterminate experimental conditions. Figure 4 is a flowchart of the experimental operation.

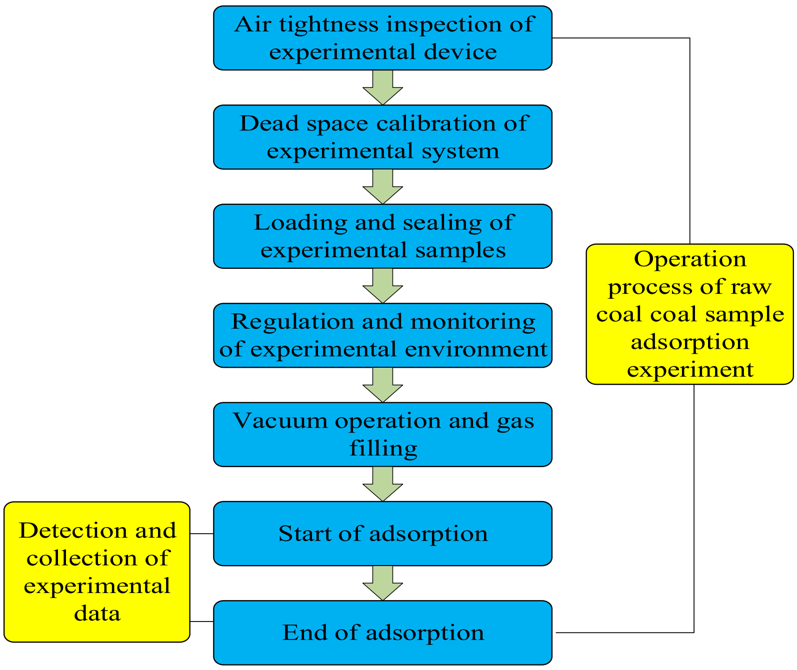

Figure 4. The flowchart of the experimental operation of the CO_2 adsorption process of the loaded coal sample under temperature conditions.

3. Results and Discussion

3.1. Variation Characteristics of Adsorption Strain of Raw Coal Samples during CO_2 Adsorption

Based on the existing experimental equipment, the experimental scheme of this paper was formulated by comprehensively considering the experimental conditions required for the research and the performance of the experimental system. This experiment was carried out to study the change characteristics of CO_2 adsorption strain of coal samples under different combination conditions through setting the temperature conditions (283.15 K/293.15 K/303.15 K), the CO_2 gas pressure conditions (3.5 MPa/4 MPa/4.5 MPa) and the axial effective stress conditions (4 MPa/4.5 MPa/5 MPa). Meanwhile, the data acquisition and analysis system were used in the experiment process to accurately record and analyze the changes of experimental conditions and the adsorption strain change law of coal sample adsorbing CO_2 under corresponding changing conditions. Considering the characteristics of CO_2 adsorption-strain variation of raw coal samples and the needs of experimental research, the stable adsorption time of the experiment was determined to be 8 h under the same experimental condition, which can not only make full use of the performance of the experimental system but also meet the requirements of this adsorption strain experiment.

Previous experimental studies on coal rock mechanics have shown that axial stress, radial stress, pore stress, loading speed and others have a great influence on the strain variation of coal samples [23]. For example, in the elastic stage of coal rock triaxial compression test, applying axial stress and radial stress and axial stress to coal sample will lead to coal sample compression and volume reduction and the main gas migration channels (such as internal pores and fissures) will be closed or contracted, which will change the adsorption and permeability of coal samples to CO_2 gas. Therefore, considering the original in-situ stress conditions of the coal reservoir, the strain state of the coal sample after stress loading was set as the initial state and the axial strain and radial strain were taken as the initial values of the adsorption strain experiment. The axial and radial expansion directions of coal samples were defined as their positive directions. The strain characteristics of CO_2 adsorption by raw coal samples under loading were analyzed and studied through the data obtained from the experiment.

The volume strain can be obtained by the following Equation:

$$\varepsilon_v = \varepsilon_x + \varepsilon_y + \varepsilon_z, \quad (1)$$

where ε_v is the volume strain, ε_x is the x-axis strain, ε_y is the y-axis strain, and ε_Z is the z-axis strain. According to the first strain invariant, the volumetric strain of the raw coal sample can be expressed as the sum of the strains in the three directions and for the cylindrical coal sample, the strains in the y-axis and z-axis directions are considered equal. This is example 2 of an Equation:

$$\varepsilon_v = \varepsilon_x + 2\varepsilon_y = \varepsilon_x + 2\varepsilon_z. \quad (2)$$

Under other conditions unchanged, effective stress is defined as the difference between average external stress and pore pressure.

$$\overline{\sigma} = \sigma - p, \quad (3)$$

where $\overline{\sigma}$ is the effective stress, σ is the average external stress, p is the pore pressure. In other words, the effective stress in a certain direction of the coal sample is equal to the difference between external stress and gas pressure in this direction.

Figure 5 shows variations of axial strain, radial strain and volume strain of coal sample with time under the conditions that axial stress is 6 MPa, radial stress is 5 MPa, CO_2 gas stress is 4.5 MPa and temperature is 283.15 K. The analysis of Figure 5 shows that the axial strain, radial strain and volume strain of coal sample in the process of CO_2 adsorption show an increasing trend with the increasing of adsorption time. Under the same adsorption time, the radial strain of the coal sample is greater than the axial strain. Moreover, with the increasing of adsorption time, the increasing rate of radial strain and axial strain decrease. The volume strain increases rapidly at the beginning of adsorption, then decreases with the increasing of adsorption time and finally tends to a stable value.

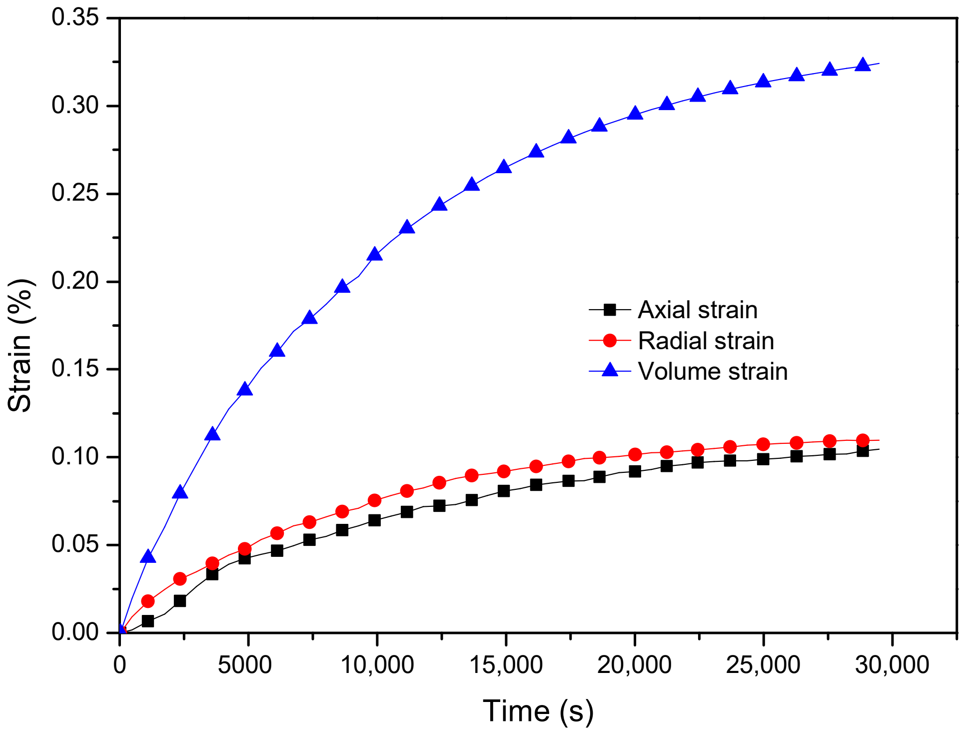

Figure 5. Variation of adsorption strain of coal sample under loading with time at 283.15 K.

Figure 6 shows the changes of axial strain, radial strain and volume strain of coal sample with time under the conditions of coal sample with axial stress of 6 MPa, radial stress of 5 MPa, CO_2 gas pressure of 4.5 MPa and temperature of 293.15 K. By changing the temperature conditions, it was obtained that the strain change of the same coal sample under different conditions as shown in Figure 6. With the increase of adsorption time, the axial strain, radial strain and volume strain of raw coal sample also showed an increasing trend. The increasing rate of radial strain and strain of coal sample at the same time point was significantly higher than that of the coal sample. Compared with the changing trend of radial strain and axial strain of the coal sample in Figure 5, it can be seen that the strain of CO_2 adsorption of raw coal sample had obvious anisotropy. Meanwhile, with the increase in temperature and the difference in the expansion effect of the coal sample in the axial and radial directions caused by the increase in temperature, the axial strain, the radial strain and the volume strain of the raw coal sample were reduced to varying degrees and the axial strain was the most obvious performance.

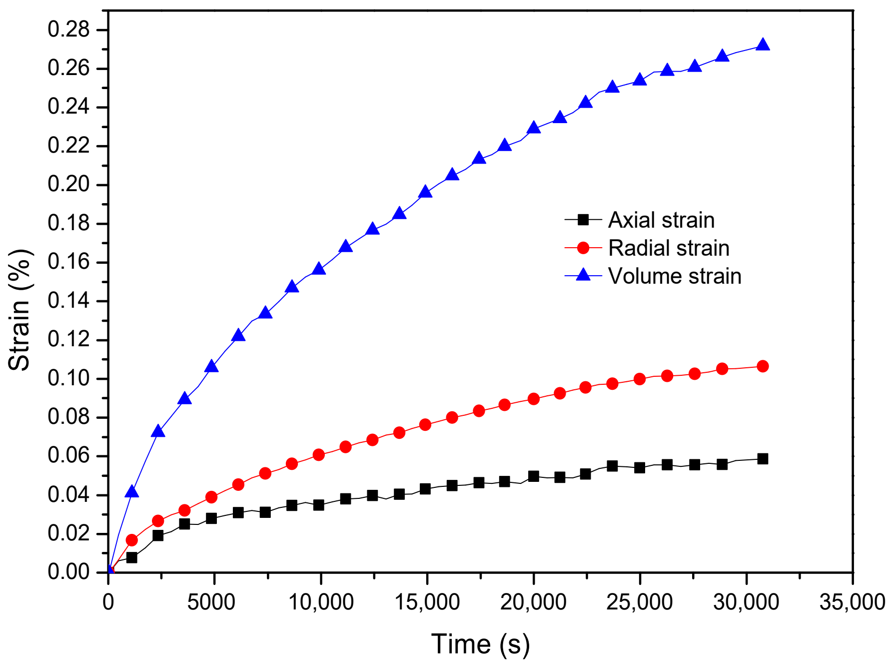

Figure 6. Variation law of CO_2 adsorption strain of loaded raw coal sample with time at 293.15 K.

3.2. Effect of Axial Effective Stress and Temperature Conditions on Strain Variation of Raw Coal Samples during CO_2 Adsorption

Under other conditions unchanged, the relationship of the axial strain of the coal sample with time was studied by only changing its axial effective stress as shown in Figure 7. Under the condition of 293.15 K and radial effective stress of 0.5 MPa, the effective axial stress of the coal sample was 1.0 MPa, 1.5 MPa and 2 MPa by changing adsorption pressure of CO_2 gas and the adsorption experiments of the loaded coal sample were respectively carried out and the axial strain variation of the loaded coal sample over time were recorded during CO_2 adsorption.

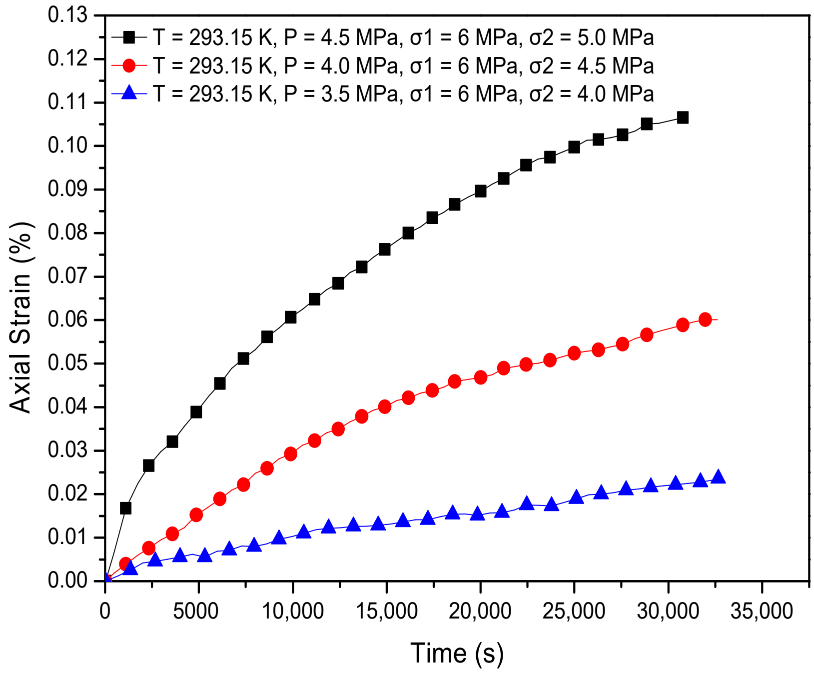

Figure 7. Variation of axial adsorption strain of coal samples under different axial effective stresses at 293.15 K.

Through comparative analysis, it can be clearly seen that the change of the axial effective stress conditions has a great impact on the adsorption performance of coal samples. And the adsorption strain shows an exponential trend with the increasing of adsorption time, Moreover, with the increasing of axial effective stress, the strain rate and the amount of strain of coal sample adsorbed CO_2 gas showed a decreasing trend.

By changing the coal sample temperature conditions (283.15 K, 293.15 K, 303.15 K) and the axial effective stress conditions (1.5 MPa, 2 MPa, 2.5 MPa), the experimental result shows that 5 sets of the relationship curves between volume strain and time of the coal sample under different axial effective stress and temperature conditions in Figure 8.

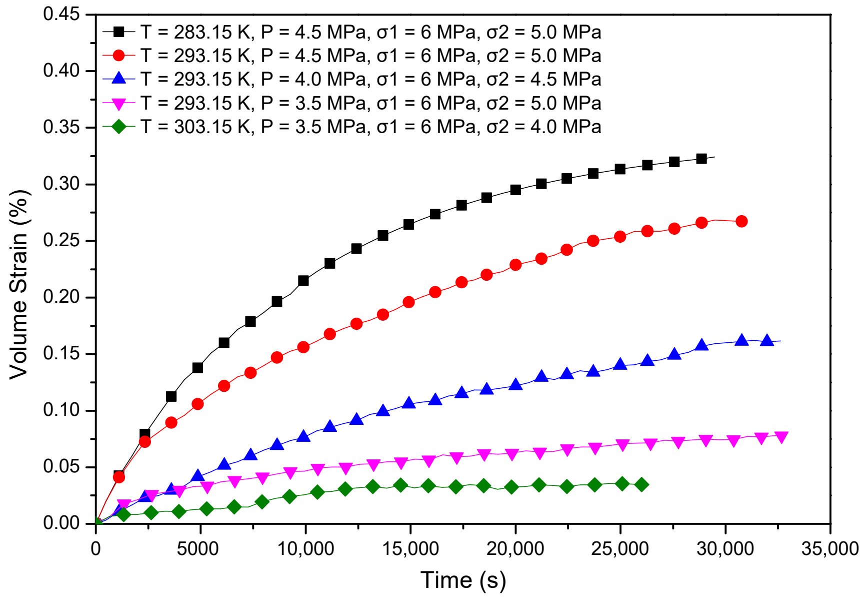

Figure 8. Variation characteristics of volume strain of coal sample with time under different temperature and effective axial stress conditions.

Figure 8 shows that the volume strain of CO_2 adsorption by the raw coal sample increases with the increasing of adsorption time and gradually tends to be flat, namely, the amount of CO_2 adsorption by coal sample tends to be saturated with the increasing of adsorption time. Under the effective radial stress unchanged and the temperature of 283.15 K and the effective axial stress of 1.5 MPa, in comparison, the volume strain of the raw coal sample is the largest and the time to reach it is the shortest at the same time point; but under the effective radial stress unchanged and the temperature of 303.15 K and the effective axial stress of 2.5 MPa, the volume strain of the coal sample is the smallest and the time to reach it is the longest at the same time point. Through the comparative analysis of Figure 8, it can be seen that: 1. The volume strain of CO_2 adsorption by coal samples increases with the increasing of adsorption time and gradually stabilizes; 2, Under other conditions unchanged, the volume strain and volume strain change of CO_2 adsorption by coal sample decrease at the same time point with the increasing of adsorption temperature, In other words, the adsorption quantity and the adsorption capacity of coal sample for CO_2 both decrease with the increase of temperature; 3, When other conditions remain unchanged, the volume strain and volume strain change of CO_2 adsorption by coal sample also show a decreasing trend with the increase of the effective axial stress at the same time point. In other words, under other conditions unchanged, the increase of temperature or the axial effective stress can both abate the CO_2 adsorption quantity of the raw coal sample and the influence on CO_2 adsorption capacity of the coal sample gradually decreases with the increasing of temperature or axial effective stress.

3.3. Discussion

In the process of CO_2-ECBM and CO_2 geological sequestration of unworkable seams, because of the influence of the expansion effect of CO_2 adsorption, it is difficult to continuously inject CO_2 into coal seams to affect the production of CO_2-ECBM and CO_2 geological sequestration. Therefore, it is very important to study the influence of geological conditions on the process of CO_2 injection into coal reservoirs, especially the study of the adsorption strain variation of the CO_2 adsorption under stress and temperature conditions. In this paper, it was carried out an experimental study on the influence of axial stress and temperature conditions on strain variation of coal sample during CO_2 adsorption. The results show that the volume strain of the raw coal sample decreases with the increasing of the temperature or the axial effective stress, namely, the increasing of the temperature or the increasing of the axial effective stress will cause the decreasing of adsorption quantity of CO_2 and the weakness of the adsorption capacity. Guan [24] carried out an experimental study on the influence of temperature conditions on the CO_2 adsorption capacity of Illinois coal. The study showed that when the experimental temperature was less than or equal to 323.15 K, the CO_2 adsorption capacity was sensitive to the temperature and linear with the increasing of the temperature but the experimental temperature was higher than 323.15 K, adsorption quantity of CO_2 was not sensitive to temperature. Zhou [25] analyzed the influence of the temperature and the pressure conditions in the process of replacing CH_4 during CO_2 injection into granular coal by the experimental method and it was concluded that the use of CO_2 to enhance coalbed methane exploitation had great advantages. At the same time, he measured the relevant parameters of coal particles in the process of CO_2 geological storage by the Helium Pycnometer method and others and believed that the increase of the temperature and the pressure cannot both evidently increase the desorption rate of adsorbed methane. Liu [26] believed that the effects of the temperature and the pressure on the CO_2 adsorption by coal reservoirs with different burial depths were mainly concentrated in the micropore range of high-grade coal and the macropore range of low-grade coal. Du [27] studied and analyzed the thermodynamic characteristics of CO_2 and CH_4 adsorption by three different grades of coal (anthracite, bituminous coal and lignite) sampled from China and considered that low temperature increased the adsorption capacity of CO_2 and CH_4 and had a greater impact on CO_2 adsorption.

Therefore, combined with the previous research and the test results in this paper, it is considered that the increase of temperature changes the gas state of CO_2 injected into the coal body results in difficult adsorption of CO_2 and reduction of the total CO_2 adsorption quantity; while the increasing of the axial effective stress results in the volume compression [28], shrinks of the internal pore and fissure structure of the coal sample and reduction of the total surface area of the coal sample for CO_2 adsorption. The increasing of the temperature or the axial effective stress will lead to the decreasing of CO_2 adsorption volume strain of coal sample, namely, it reduces the total CO_2 adsorption quantity of coal sample. In the process of enhanced coalbed methane and CO_2 Geological sequestration, it is very important that the influence of temperature and effective stress condition on CO_2 adsorption of coal samples. And the temperature conditions of CO_2 adsorption not only have an important influence on the phase state of CO_2 but also have a coupling effect with geological reservoirs to affect the migration process of CO_2 in reverse. Therefore, it is not only an important supplement to the existing theory but also a great significance to the practical engineering application to study the adsorption-desorption and seepage mechanism of multiphase CO_2 (CO_2/LCO_2/SCO_2) injection process and the influence on the coupling of geological reservoirs and their migration channels. But, Due to the limitation of experimental conditions, this paper only studied and analyzed the influence of temperature and axial effective stress on the CO_2 adsorption process of raw coal samples at the temperature range of 283.15–303.15 K and showed some relatively simple experimental results. It is the focus of our next work to study the strain change law of the raw coal sample during CO_2 adsorption under a wider range of temperature and the stress conditions and how the coupling effect of temperature and stress conditions changes the law of adsorption-desorption and permeability of coal reservoirs, as well as the analysis of the primary and secondary relationship between temperature and stress in this process.

4. Conclusions

In this paper, the laboratory simulation experiments are conducted to study the strain change law of raw coal samples during CO_2 adsorption under the conditions of temperature and axial effective stress. Despite the limitations of the experiment, the experimental results show that the temperature and the axial effective stress conditions do have a clear impact on the strain of the raw coal sample for CO_2 adsorption. According to the experimental results, we have the following conclusions:

(1) With the increase of CO_2 adsorption time, the axial strain and the radial strain of the coal sample simultaneously increase and the increasing rate shows the trend of first fast and then slow. At the same time, the axial strain and the radial strain of the coal sample show obvious anisotropy in the CO_2 adsorption process.

(2) Under other conditions unchanged, the greater the axial effective stress, the smaller the axial strain growth rate and the maximum axial deformation of the loaded raw coal sample during the process of CO_2 adsorption, which shows that the axial stress increases to close or compress the pore and fissure structure in the raw coal sample and result in the decrease of CO_2 adsorption quantity, which further affects the axial deformation of the raw coal samples adsorbing CO_2.

(3) Under other conditions unchanged, the increase of temperature or the axial effective stress can both reduce the CO_2 adsorption quantity of coal samples and the influence on CO_2 adsorption capacity of coal samples gradually decreases with the increase of temperature or axial effective stress. Namely, the temperature and stress conditions have a very important impact on the CO_2 migration process and engineering effect in the enhanced coalbed methane and theCO_2 Geological sequestration.

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