

## Article

# Numerical Simulation of the Effect of Injected CO<sub>2</sub> Temperature and Pressure on CO<sub>2</sub>-Enhanced Coalbed Methane

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**Abstract:** The injection of CO<sub>2</sub> to displace CH<sub>4</sub> in coal seams is an effective method to exploit coalbed methane (CBM), for which the CO<sub>2</sub> injection temperature and pressure are important influential factors. We performed simulations, using COMSOL Multiphysics to determine the effect of CO<sub>2</sub> injection temperature and pressure on CO<sub>2</sub>-enhanced coalbed methane (CO<sub>2</sub>-ECBM) recovery, according to adsorption/desorption, seepage, and diffusion of binary gas (CO<sub>2</sub> and CH<sub>4</sub>) in the coal seam, and derive a thermal-hydraulic-mechanical coupling equation of CO<sub>2</sub>-ECBM. The simulation results show that, as CO<sub>2</sub> injection pressure in CO<sub>2</sub>-ECBM increases, the molar concentration and displacement time of CH<sub>4</sub> in the coal seam significantly decrease. With increasing injection temperature, the binary gas adsorption capacity in the coal seam decreases, and CO<sub>2</sub> reserves and CH<sub>4</sub> production decrease. High temperatures are therefore not conducive for CH<sub>4</sub> production.

**Keywords:** coalbed methane; CO<sub>2</sub>-ECBM; thermal-hydraulic-mechanical; temperature; pressure

## 1. Introduction

Coalbed methane (CBM) in coal seams is usually stored as free gas in cracks and pores and adsorbed gas on organic surfaces [1]. CBM production not only eliminates the threat of hazardous coal mine production and prevents gas over-limits, but also offers economic benefits [2,3]. CO<sub>2</sub> is a major greenhouse gas and the biggest contributor to climate change [4–8], because its adsorption capacity is higher than that of CH<sub>4</sub>. CO<sub>2</sub> displacement can be adopted for CBM recovery [9]. CO<sub>2</sub>-enhanced coalbed methane (CO<sub>2</sub>-ECBM) technology involves the injection of CO<sub>2</sub> into a coal seam rich in CBM, to sequester CH<sub>4</sub>, promotes clean green energy, and is widely used in the production of deep ultra-low permeability coal seams [10–14].

Dell reported that CH<sub>4</sub> could be effectively extracted from crushed coal by injecting flowing CO<sub>2</sub> at ambient temperature [15]. Gentzis injected CO<sub>2</sub> waste from CBM power plants into a coal seam and produced more CH<sub>4</sub>, obtaining a competitive adsorption CH<sub>4</sub>:CO<sub>2</sub> ratio of approximately 2:1 [16]. Robertson and Christiansen found that H<sub>2</sub>S, CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub> expand to different degrees in coal seams, and the strain caused by CO<sub>2</sub> adsorption was the largest [17]. Charrière conducted CH<sub>4</sub> and CO<sub>2</sub> adsorption experiments on coal at different temperatures and found that the equilibrium time of coal adsorption of CH<sub>4</sub> and CO<sub>2</sub> is negatively correlated with pressure and temperature and that matrix expansion leads to a decrease of pore width within coal seam fractures [18]. This leads to a

significant decrease in permeability. The degree of coal expansion caused by  $\text{CO}_2$  is greater than that caused by  $\text{CH}_4$  [19]. Wei established a multi-component gas diffusion kinetic model based on a Bidisperse diffusion mechanism and Maxwell–Stefan (MS) diffusion theory to verify model effectiveness under pure gas diffusion conditions [20]. Fujioka studied the feasibility of storing  $\text{CO}_2$  underground while extracting  $\text{CH}_4$  from coal seams and showed that  $\text{CO}_2$  injection can increase gas production [21]. The results of Luo showed that vertical permeability heterogeneity can improve the transport capacity of  $\text{CO}_2$  to a production well [22]. Vishal used numerical simulations to study the production effect of coal with different sorption times under  $\text{CO}_2$  action. Their results indicate that the  $\text{CO}_2$  injection capacity of coal with high sorption time is higher than that of coal with low sorption time [23]. Wang established a fully coupled gas flow model based on double permeability diffusion, adsorption strain, and geomechanics and showed that the double-pore diffusion model better describes the diffusion process than the single-pore diffusion model [24]. Fan established the hydraulic–mechanical–thermal coupling model of  $\text{CO}_2$ -ECBM, considering the diffusion of  $\text{CO}_2$  and  $\text{CH}_4$  gas and non-isothermal initial temperature absorption of coal, and simulated the effect of injection pressure and initial reservoir temperature on  $\text{CO}_2$ -ECBM [25]. Fang established a fully coupled equation of gas diffusion, adsorption, seepage, and heat transfer and simulated the displacement process and effective influence radius of injected  $\text{CO}_2$  under different pressure and temperature conditions [26].

The abovementioned studies on  $\text{CO}_2$ -ECBM established the developmental foundation CBM displacement by  $\text{CO}_2$ . However, most scholars have assumed a single temperature or pressure parameter in the study of numerical simulation of  $\text{CO}_2$ -ECBM, and most of them set the coal seam as a homogeneous body. In this paper, we established the THM coupling equation of  $\text{CO}_2$ -ECBM for coal seams with non-uniform porosity and that are non-isothermal, and non-isobaric adsorption of binary gas is considered while combining percolation and diffusion. To study the effects on gas recovery and coal seam permeability, we used COMSOL Multiphysics to simulate the process of injecting  $\text{CO}_2$  with different pressures and temperatures to displace coalbed methane.

## 2. Theory

### 2.1. Desorption–Seepage–Diffusion Principle of $\text{CO}_2$ Displacement of $\text{CH}_4$

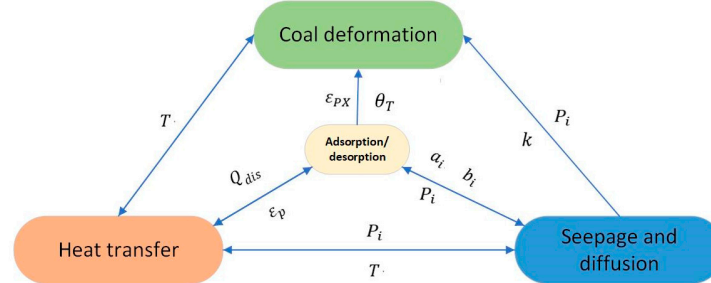
The  $\text{CO}_2$  adsorption capacity of coal seams is greater than that of  $\text{CH}_4$ . After injecting  $\text{CO}_2$  into a coal seam, adsorption competes with  $\text{CH}_4$  and some of the latter is simultaneously displaced. The injection of  $\text{CO}_2$  increases the energy of the coal seam and produces a partial pressure effect on the adsorption of  $\text{CH}_4$ , which reduces the adsorption pressure of  $\text{CH}_4$  and promotes its desorption. After desorption,  $\text{CO}_2$  changes from an adsorbed state to a free state. Gas molecules diffuse under a concentration and pressure gradient, migrating from higher concentration to lower concentration and then to the wellbore of the production well.

### 2.2. THM Coupling Equation of $\text{CO}_2$ Displacement CBM

The process of  $\text{CO}_2$  displacement of CBM mainly involves THM coupling, as shown in Figure 1. We therefore establish a THM-coupling model of deformation, heat transfer, gas adsorption/desorption, seepage, and diffusion of coal with a heterogeneous porous medium. The field equation is based on the following assumptions:

- (1) The coal seam is a heterogeneous porous medium;
- (2) The binary gas adsorption and desorption models conform to the Langmuir equation [27];
- (3) The influence of water and vapor on gas transport is not considered [28];
- (4) The free-state carbon dioxide and methane gas in the coal seam obey the ideal gas state equation (ignoring the influence of gas compression coefficient and temperature on gas viscosity) [25–27,29–31];

- (5) The migration mode of binary gas in the coal seam pores obeys Fick diffusion law, the free binary gas transport in coal seam obeys Darcy's law, and binary gas mass exchange occurs between diffusion and seepage [16,32];
- (6) The initial state of the coal seam's only free adsorption state of CBM sets CO<sub>2</sub> content to 0 with binary gas in the boundary around the coal seam as no flux.



**Figure 1.** Schematic diagram of the full coupling of THM for CO<sub>2</sub> displacement CBM.

### 2.2.1. Stress–Strain Equation

The coal seam is a heterogeneous porous medium model. The deformation field of coal seam is affected by pore pressure, temperature, and matrix expansion caused by gas adsorption and desorption. The stress is expressed as the strain equation [25,33–36]:

$$\varepsilon_{ij} = \frac{1}{2G} \delta_{ij} - \left( \frac{1}{6G} - \frac{1}{9K} \right) \sigma_{kk} \delta_{ij} + \varepsilon_{py} \delta_{ij} + \varepsilon_T \delta_{ij} + \varepsilon_{px} \delta_{ij} \quad (1)$$

where  $G$  is the shear modulus (Pa);  $K$  is the bulk modulus of coal (Pa);  $\varepsilon_{py}$  is gas strain;  $\varepsilon_T$  is thermal expansion strain; and  $\varepsilon_{px}$  is the strain caused by gas pressure. The equation is as follows:

$$\begin{cases} G = \frac{E}{2(1+\nu)} \\ K = \frac{E}{3(1-2\nu)} \\ \varepsilon_{py} = -\frac{\alpha \Delta P}{3K_s} \\ \varepsilon_T = \frac{\beta \Delta T}{3} \\ \varepsilon_{px} = \frac{2\rho_c R T a_i}{3V_m K} \ln(1 + \sum_{i=1}^2 b_i P_i) \end{cases} \quad (2)$$

where  $E$  is the Young's modulus (Pa);  $\nu$  is the Poisson's ratio;  $\beta$  is the thermal expansion coefficient (K<sup>-1</sup>);  $V_m$  is the molar constant of gas;  $R$  is the universal gas constant (J/mol·k);  $\delta_{ij}$  is the Kronecker function;  $a_i$  is the Langmuir volume constant (m<sup>3</sup>/kg);  $b_i$  is the Langmuir pressure constant (Pa<sup>-1</sup>);  $P_i$  is the gas pressure (MPa);  $\rho_c$  is the density of coal (kg/m<sup>3</sup>); and  $T$  is the temperature of the coal (K).

The strain and displacement components satisfy the Cauchy equation [28,37]:

$$\varepsilon_{ij} = \frac{1}{2} (u_{ij} + u_{ji}) \quad (3)$$

where  $\varepsilon_{ij}$  is the strain component, and  $u_{ij}$  is the displacement component.

The equilibrium differential equation of coal seam is defined as the Navier–Stokes equation [28,37]:

$$\sigma_{ij,j} + F_i = 0 \quad (4)$$

The modified equilibrium differential equation is as follows [38]:

$$\sigma'_{ij,j} + (\alpha P \sigma_{ij})_{,j} + F_i = 0 \quad (5)$$

Combined with Equations (1)–(5), the coal stress equation is as follows:

$$Gu_{i,jj} + \frac{G}{1-2\nu} u_{j,ji} + \theta_{py} \Delta P_{,i} + \theta_T \Delta T_{,i} + \theta_{PX} a_i T \left[ \ln(1 + \sum_{i=1}^2 b_i P_i) \right]_{,i} + \alpha P_{,i} + F_i = 0 \quad (6)$$

where  $\theta_{py}$  is the stress coefficient caused by gas pressure,  $\theta_T$  is the coefficient of thermal stress,  $\theta_{PX}$  is the stress coefficient caused by gas adsorption, and  $\alpha$  is the Biot coefficient. The equation is as follows:

$$\begin{cases} \theta_{py} = \frac{3\lambda - 2G}{3K} \\ \theta_T = \frac{3\lambda + 2G}{3} \beta \\ \theta_{PX} = \frac{2\rho_c R(3\lambda + 2G)}{3V_m K} \\ \alpha = 1 - K / K_s \\ \lambda = \frac{Ev}{(1+\nu)(1-2\nu)} \end{cases} \quad (7)$$

where  $K_s$  is bulk modulus of coal skeleton, and  $\sigma_{ij}$  is stress tensor ( $i, j = 1, 2$ ).

### 2.2.2. Coupling Equation of Permeation and Diffusion of Binary Gas

Gas seepage in coal seams is driven by a pressure gradient and gas gravity is ignored. The gas seepage velocity expression in coal is derived from Darcy's law [25]:

$$q_g = -\frac{KRT}{\mu_i} \nabla c_i \quad (8)$$

where  $q_g$  is seepage velocity ( $\text{m}^3/\text{s}$ );  $\nabla = \left[ \frac{\partial}{\partial x}, \frac{\partial}{\partial y}, \frac{\partial}{\partial z} \right]$ ; and  $\mu_i$  is viscosity coefficient of single phase.

Gas diffusion follows Fick's law. Its diffusion flux is expressed as follows [39]:

$$J_i = -D_i \nabla c_i \quad (9)$$

where  $J_i$  is the diffusion component of single phase;  $D_i$  is the diffusion coefficient of single phase ( $\text{m}^2/\text{s}$ ).

$\text{CO}_2$  is injected into coal seams containing  $\text{CH}_4$  driven by a pressure gradient, and the flow of binary gas in coal seam conforms to the convection–diffusion equation [25]:

$$\frac{\partial(m_i)}{\partial t} + \nabla \cdot \left( -\frac{kRT}{\mu_i} \nabla c_i \rho_i \right) + \nabla \cdot (-D_i \phi \nabla c_i) = Q_s \quad (10)$$

where  $i$  is the single component gas;  $i=1$  is  $\text{CH}_4$ ;  $i=2$  is  $\text{CO}_2$ ;  $m_i$  is the gas mass of each component ( $\text{kg}$ );  $c_i$  is the molar concentration of a single component ( $\text{mol}/\text{m}^3$ );  $\rho_i$  is the density of a single component ( $\text{kg}/\text{m}^3$ );  $k$  is permeability ( $\text{m}^2$ );  $\phi$  is porosity; and  $Q_s$  is the source term ( $\text{kg}/(\text{m}^3 \cdot \text{s})$ ).

For the ideal state equation of binary mixed gas, the molar concentration of each gas is expressed as follows [24]:

$$c_i = \frac{P_i}{RT} \quad (11)$$

CO<sub>2</sub> and CH<sub>4</sub> gases exist simultaneously in the coal seam during CO<sub>2</sub> displacement. The mass of the gas is the sum of adsorbed and free gases considering the influence of temperature on the adsorption capacity. The Langmuir adsorption equation can be used to represent the adsorption mass as follows:

$$m_i = \rho_c \rho_a \frac{a_i b_i c_i RT}{1 + \sum_{i=1}^2 b_i P_i} + \phi M_i c_i \quad (12)$$

Combined with Equations (8)–(12), the binary gas seepage and diffusion equation can be obtained as follows:

$$\frac{\partial \left( \rho_c \rho_a \frac{a_i b_i c_i RT}{1 + \sum_{i=1}^2 b_i P_i} + \phi M_i c_i \right)}{\partial t} + \nabla \cdot \left( -\frac{kRT}{\mu_i} \nabla c_i \rho_i \right) + \nabla \cdot (-D_i \phi \nabla c_i) = Q_s \quad (13)$$

### 2.2.3. Temperature Field Equation of Binary Gas Flow

The system is treated as the thermal equilibrium state, and the coal seam is set as a porous medium of linear thermoelastic material [25]. Deformation work and heat applied to the coal seam are equal to the sum of its kinetic and internal energies. Because deformation of the coal seam is small and reversible, the kinetic energy can be ignored. The energy balance equation of the coal seam is then as follows:

$$\delta Q_H = dU - \delta_{ij} d\epsilon_{ij} = \rho_c C_V dT + T \frac{\partial \delta_{ij}}{\partial t} d\epsilon_{ij} \quad (14)$$

where  $dQ_H$  is the heat source for thermal expansion, volumetric deformation, and adsorption deformation caused by temperature changes;  $dU$  is the internal energy per unit volume; and  $C_V$  is specific heat at constant volume of coal seam.

The partial derivative of the thermoelastic physical equation is obtained:

$$\frac{\partial \sigma_{ij}}{\partial t} = \frac{\partial \left[ -Q_T \Delta T \delta_{ij} + Q_{PX} \Delta P \delta_{ij} + Q_{PY} a_i T \left[ \ln(1 + \sum_{i=1}^2 b_i P_i) \right] \delta_{ij} \right]}{\partial t} \quad (15)$$

Ignoring the influence of water and ash content on the coal seam, the heat source in the coal seam is mainly composed of energy absorbed by gas adsorption, release, and desorption. The heat generated by coal deformation, the gas flow temperature field equation, is as follows:

$$dQ_H = \eta \nabla^2 T + Q_{dis} \quad (16)$$

where  $\eta$  is thermal conductivity of coal;  $\eta \Delta^2 T$  is the change of heat flow into and out of unit volume by heat conduction of gas in unit time; and  $Q_{dis}$  is differential heat source.

Because binary gas desorption of the coal seam is a reversible adsorption process, gas desorption in the coal seam changes from an adsorption state to a free state. The expressions of heat absorbed during CH<sub>4</sub> desorption in a coal seam and heat released during CO<sub>2</sub> adsorption are as follows:

$$Q_{dis} = \frac{\partial \left[ \frac{a_i b_i P_i \rho_a \rho_i RT}{M_i (1 + \sum_{i=1}^2 b_i P_i)} \ln \left[ \frac{\phi M_i (1 + \sum_{i=1}^2 b_i P_i)}{a_i b_i \rho_a \rho_i RT} \right] \right]}{\partial t} \quad (17)$$

By combining Equations (14)–(17), the temperature field control equation of binary gas is obtained as follows:

$$\eta \nabla^2 T + Q_{dis} = \rho_c C_V \frac{\partial T}{\partial t} + Q_T \Delta T \delta_{ij} \frac{\partial e}{\partial t} + Q_{PX} a_i T \left[ \ln(1 + \sum_{i=1}^2 b_i P_i) \right] \frac{\partial e}{\partial t} \quad (18)$$

#### 2.2.4. Porosity and Permeability

The adsorption and desorption of gas in the seepage and diffusion field, compression of the stress field on the coal seam, and thermal expansion of the temperature field are transformed into the influence on porosity, which is defined as follows [40]:

$$\varphi = 1 - \frac{1 - \varphi_0}{1 + e} \left( 1 + \frac{\Delta V_S}{V_{S0}} \right) \quad (19)$$

where  $\varphi_0$  is the initial porosity of coal;  $e$  is the volumetric strain of coal;  $V_{S0}$  is the initial volume of coal skeleton;  $\Delta V_S$  is the coal skeleton volume changes under the comprehensive action of pore pressure compression, thermal expansion, and gas absorption expansion.

The effects of gas pressure and coal temperature on porosity are considered:

$$\varphi = 1 - \frac{1 - \varphi_0}{1 + e} \left( 1 + \frac{\Delta V_{SP}}{V_{S0}} + \frac{\Delta V_{ST}}{V_{S0}} \right) \quad (20)$$

where  $\Delta V_{SP}$  is bulk expansion and deformation of coal caused by pressure;  $\Delta V_{ST}$  is bulk expansion and deformation of coal caused by temperature change, and the equation is as follows:

$$\begin{cases} \frac{\Delta V_{SP}}{V_{S0}} = -\frac{\Delta P}{K_S} \\ \frac{\Delta V_{ST}}{V_{S0}} = \beta \Delta T \end{cases} \quad (21)$$

Volumetric deformation resulting from gas adsorption and desorption is given as follows [41]:

$$\varepsilon_p = \frac{2\rho_c R T a_i}{3V_m K} \ln(1 + \sum_{i=1}^2 b_i P_i) \quad (22)$$

The porosity equation can be obtained by integrating the porosity factors:

$$\varphi = 1 - \frac{1 - \varphi_0}{1 + e} \left( 1 - K_Y \Delta P + \beta \Delta T + \frac{\frac{2\rho_c R T a_i}{3V_m K} \ln(1 + \sum_{i=1}^2 b_i P_i)}{1 - \varphi_0} \right) \quad (23)$$

The relationship between porosity, permeability, and particle size distribution in porous media is as follows [29,33]:

$$k = \frac{d_e^2 \varphi^3}{72(1 - \varphi)^2} \quad (24)$$

where  $d_e$  is the effective diameter of particle. According to Equation (22), we obtain:

$$\frac{k}{k_0} = \left( \frac{\varphi_0}{\varphi} \right)^3 \left( \frac{1 - \varphi_0}{1 - \varphi} \right)^2 \quad (25)$$

The second term on the right tends to be consistent when porosity is substantially lower than 1. The relationship between porosity and permeability is as follows [42,43]:

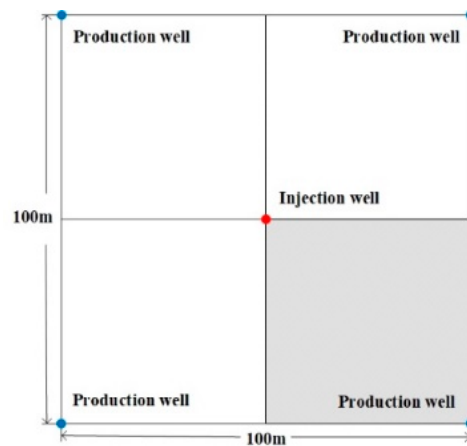
$$k = k_0 \left( \frac{\varphi}{\varphi_0} \right)^3 \quad (26)$$

where  $k_0$  is the initial permeability ( $\text{m}^2$ ), and coal seam permeability is as follows:

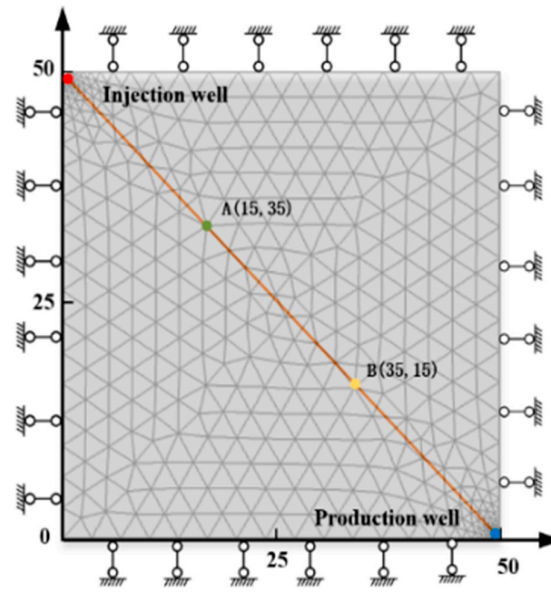
$$k = k_0 \left( \frac{1 - \frac{1 - \varphi_0}{1 + e} \left( 1 + \frac{\Delta V_{SP}}{V_{S0}} + \frac{\Delta V_{ST}}{V_{S0}} \right)}{\varphi_0} \right)^3 \quad (27)$$

### 3. Geometric Model and Solution Conditions

We simplify the CBM reservoir in Northern Sichuan (Qinshui Basin, China) into a two-dimensional model, according to the production block of  $\text{CO}_2$  displacement of  $\text{CH}_4$  and ignoring the coal seam thickness. The geometric model simplify is simplified to a two-dimensional model of a square area with a side length of 100 m. Considering the symmetry, the 1/4 of the area is selected as a numerical simulation area. As shown in Figure 2, COMSOL Multiphysics software was used to establish a coal seam model with a size of  $50 \times 50$  m. A 1 m diameter production well is located at the bottom right of the model and the pressure is set to 1 atm (0.1 MPa). An injection well with a 1 m diameter is located in the upper left of the model. A triangular mesh was used to divide into 25,504 cells. To facilitate the observation of simulation results, A (15, 35) and B (35, 15) were set as simulation monitoring points of the model. The initial pressure and temperature of the coal seam are 2 MPa and 273 K, respectively. The coal seam and well boundaries are fixed constraints. The initial porosity of each point is  $\varphi_{0(x,y)}$  and the model parameters are listed in Table 1.



(a)



(b)

(c)

**Figure 2.** Numerical model of CO<sub>2</sub> displacement of coalbed methane: (a) geological model of enhancing CBM recovery by injecting CO<sub>2</sub> with heat injection; (b) numerical simulation model for CO<sub>2</sub>-ECBM; and (c) initial porosity of CO<sub>2</sub>-ECBM.

**Table 1.** Numerical simulation parameter [25,26,28].

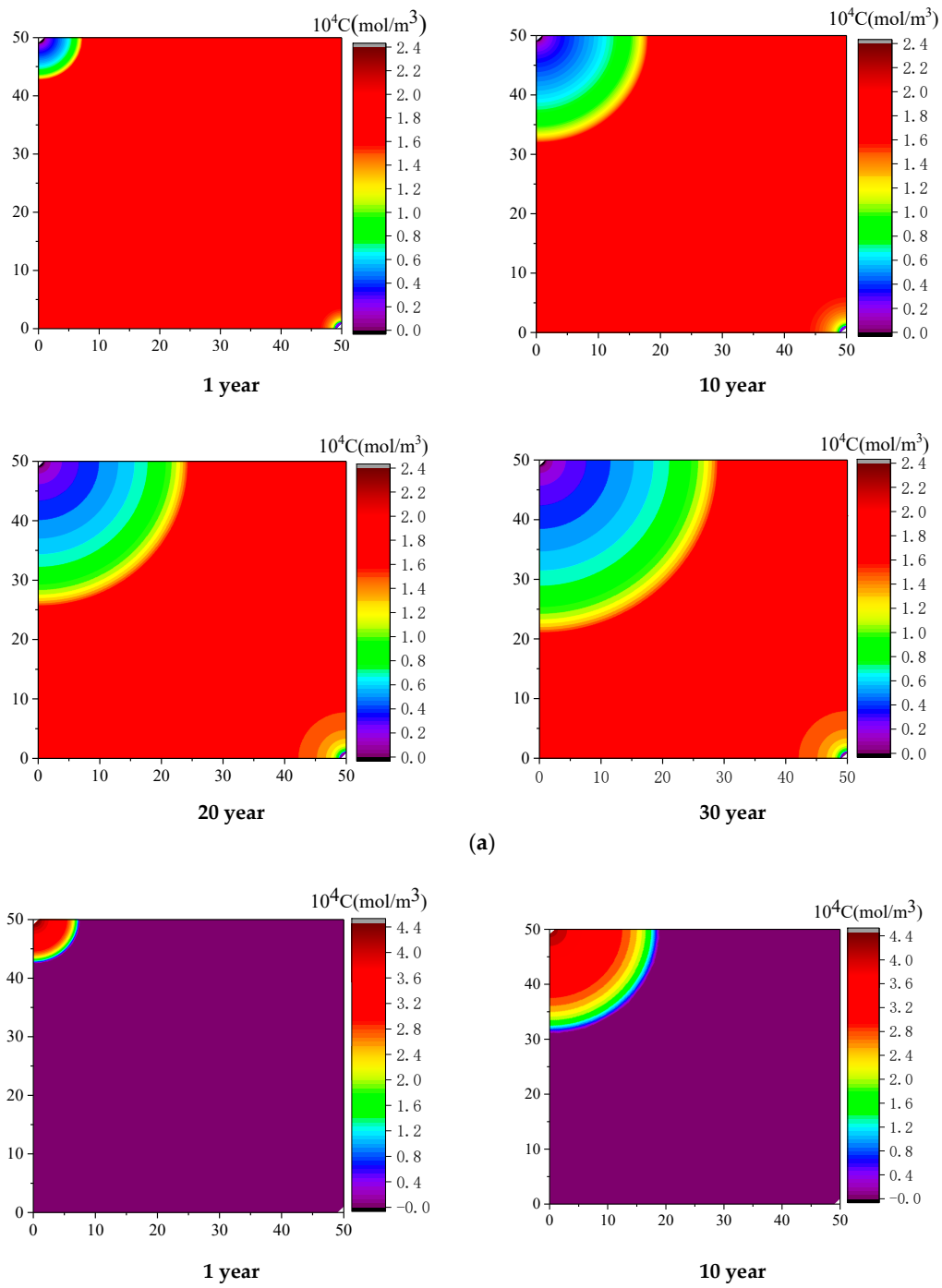
| Variable | Parameter                                            | Value                 |
|----------|------------------------------------------------------|-----------------------|
| $P_0$    | Gas pressure under standard conditions (MPa)         | 2                     |
| $T_0$    | Initial temperature of coal seam (K)                 | 273                   |
| $\rho_c$ | The density of coal (kg/m <sup>3</sup> )             | 1350                  |
| $M_1$    | CH <sub>4</sub> molar mass (kg/mol)                  | 0.016                 |
| $M_2$    | CO <sub>2</sub> molar mass (kg/mol)                  | 0.044                 |
| $\mu_1$  | CH <sub>4</sub> dynamic viscosity coefficient (Pa·s) | $1.03 \times 10^{-5}$ |
| $\mu_2$  | CO <sub>2</sub> dynamic viscosity coefficient (Pa·s) | $1.38 \times 10^{-5}$ |

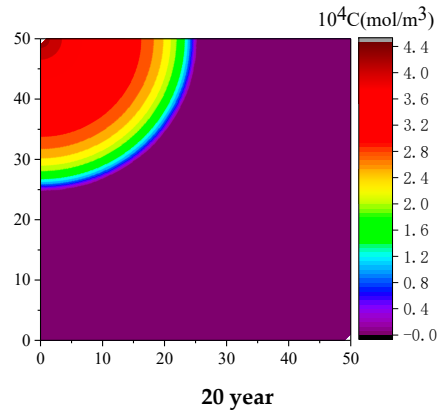
|          |                                                                        |                      |
|----------|------------------------------------------------------------------------|----------------------|
| $\rho_1$ | CH <sub>4</sub> density under standard conditions (kg/m <sup>3</sup> ) | 0.717                |
| $\nu$    | Poisson's ratio                                                        | 0.35                 |
| $E$      | Young's modulus of coal (MPa)                                          | 2713                 |
| $R$      | Universal gas constant (J/(mol·K))                                     | 8.314                |
| $\sigma$ | Thermal conductivity (W/(m·K))                                         | 0.478                |
| $\beta$  | Thermal expansion coefficient (K <sup>-1</sup> )                       | $2.4 \times 10^{-5}$ |
| $C_p$    | Heat capacity at constant stress (J/(kg·K))                            | 1000                 |

## 4. Results

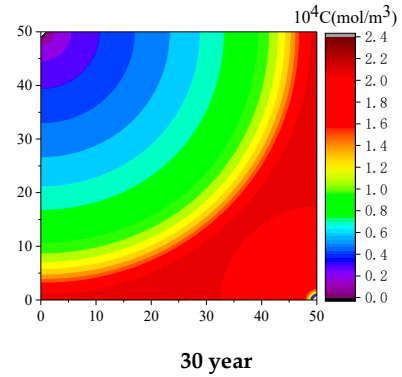
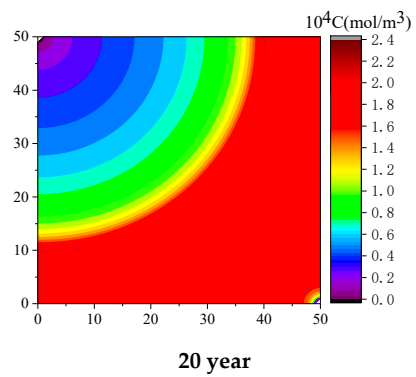
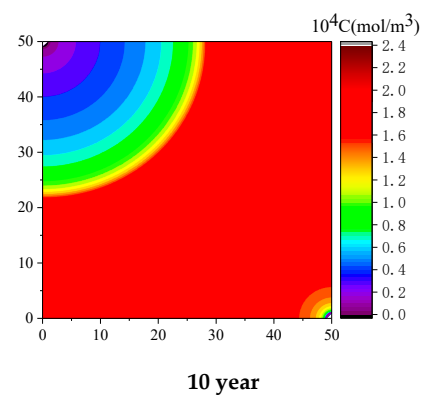
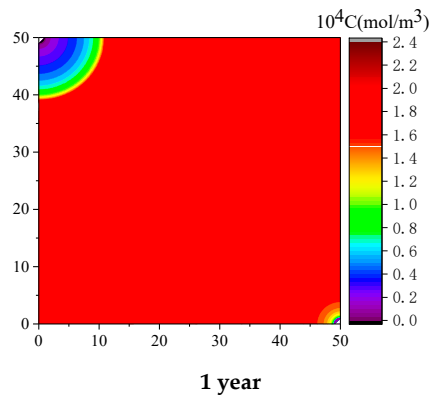
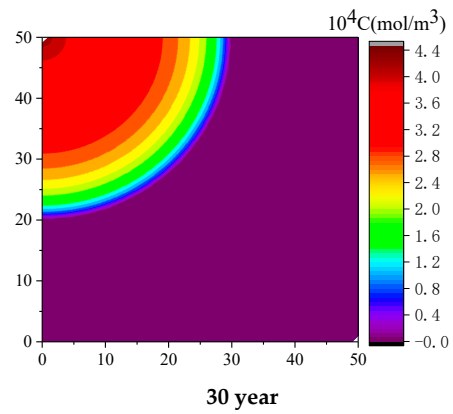
### 4.1. Effect of Injected CO<sub>2</sub> Pressure on CO<sub>2</sub>-ECBM

According to [25,26], hydraulic-mechanical-thermal coupled model of CO<sub>2</sub>-ECBM can effectively reveal the influence of CO<sub>2</sub> storage on CH<sub>4</sub> production. Therefore, we investigate the distribution rule of CO<sub>2</sub> and CH<sub>4</sub> molar concentrations in CBM over a period of 30 years to study the effect of CO<sub>2</sub> injection pressure on CO<sub>2</sub>-ECBM at constant temperature and variable pressure. The injected CO<sub>2</sub> pressures are 4, 6, and 8 MPa, and the initial temperature of the coal seam is 273 K. Figure 3 shows the migration relationship of CH<sub>4</sub> and CO<sub>2</sub> molar concentration with time under injected CO<sub>2</sub> of variable pressure.

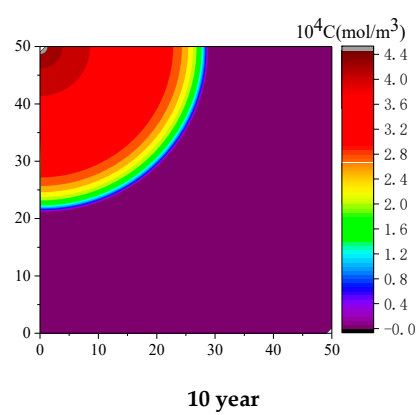
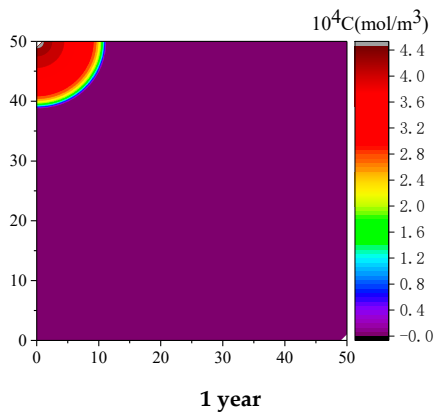


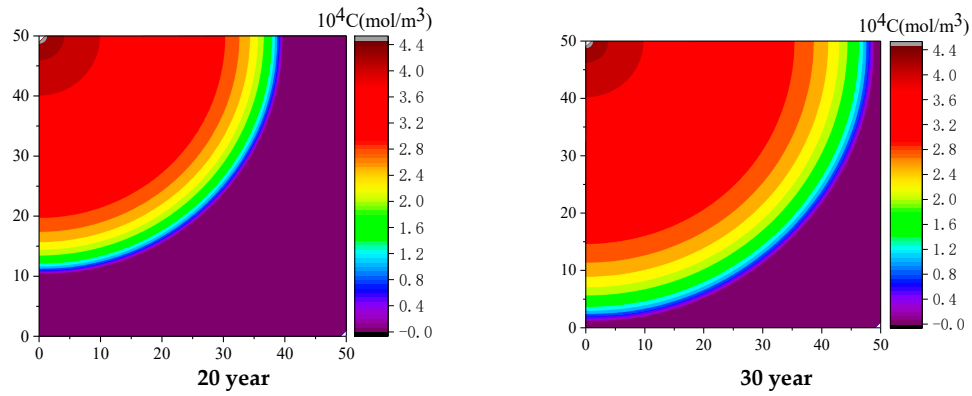


(b)

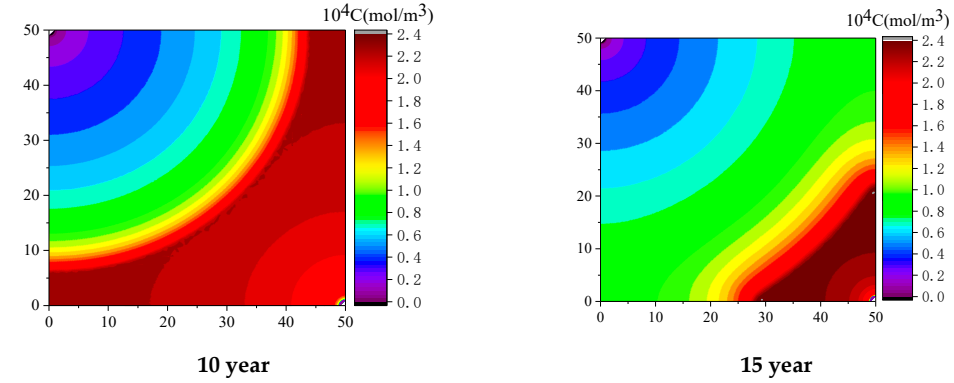
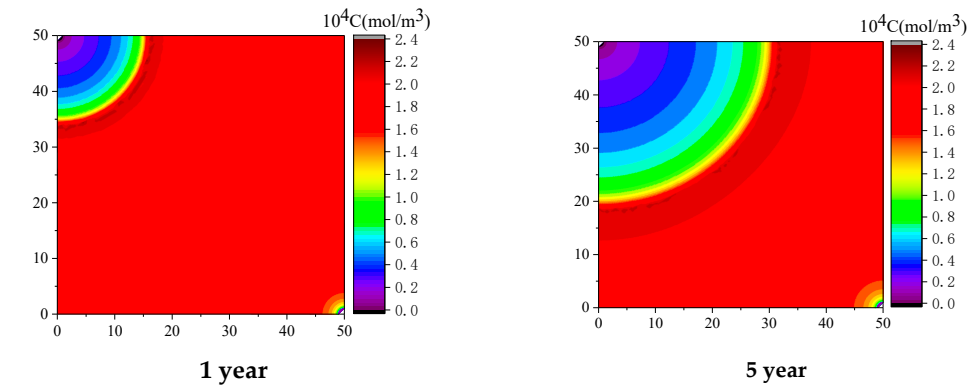


(c)

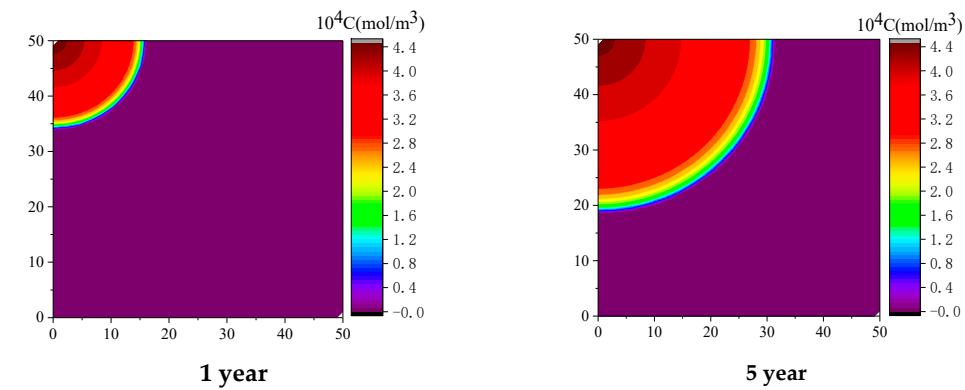


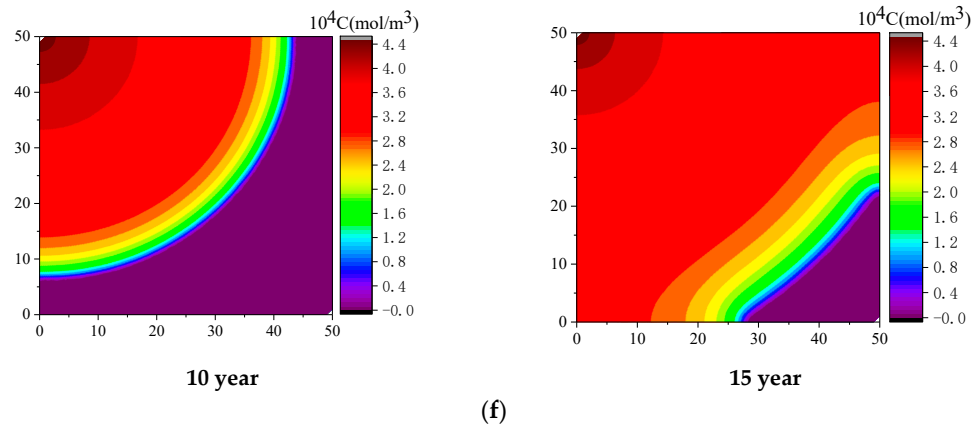


(d)



(e)

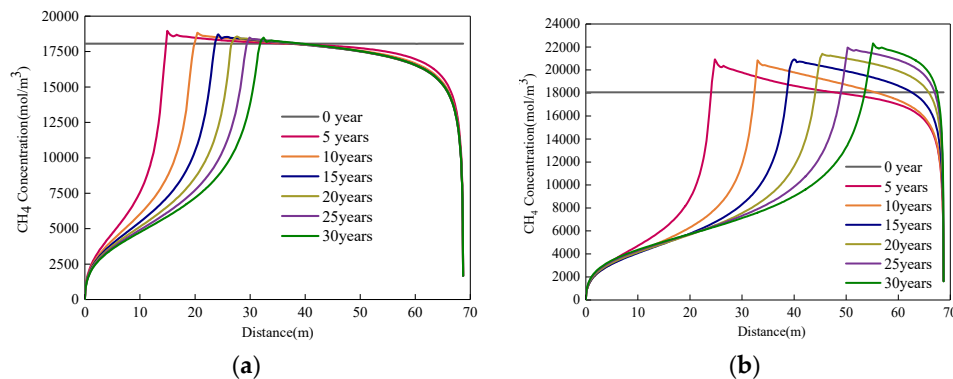


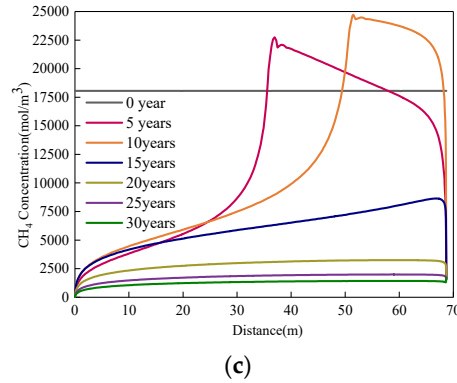


**Figure 3.** Distribution of CH<sub>4</sub> molar concentration and CO<sub>2</sub> molar concentration under different injection CO<sub>2</sub> pressures: (a) distribution of CH<sub>4</sub> molar concentration at 4 MPa injection pressure; (b) distribution of CO<sub>2</sub> molar concentration at 4 MPa injection pressure; (c) distribution of CH<sub>4</sub> molar concentration at 6 MPa injection pressure; (d) distribution of CO<sub>2</sub> molar concentration at 6 MPa injection pressure; (e) distribution of CH<sub>4</sub> molar concentration at 8 MPa injection pressure; and (f) distribution of CO<sub>2</sub> molar concentration at 8 MPa injection pressure.

Figure 3 shows the migration of CH<sub>4</sub> and CO<sub>2</sub> molar concentration with time under different injected CO<sub>2</sub> pressures. Figure 3a,c,e shows CH<sub>4</sub> molar concentration when the injection pressure is 4, 6, and 8 MPa, respectively. It can be seen that the CO<sub>2</sub> injection pressure significantly affects the displacement and migration rate of CH<sub>4</sub>. Moreover, higher CO<sub>2</sub> injection pressures are associated with faster CH<sub>4</sub> migration rates and lower molar concentrations. Figure 3b–f shows the molar concentration of CO<sub>2</sub> when the injection pressure is 4, 6, and 8 MPa, respectively. Increasing the injection pressure and time leads to an increase in influence radius and molar CO<sub>2</sub> concentration in the coal seam. After 30 years of production, when the injection pressure is 4 and 6 MPa, the influence radius of CO<sub>2</sub> is 32 and 55 m, respectively. CO<sub>2</sub> reaches the producing well 17 years after production at an injection pressure of 8 MPa. The results are basically consistent with [25,26].

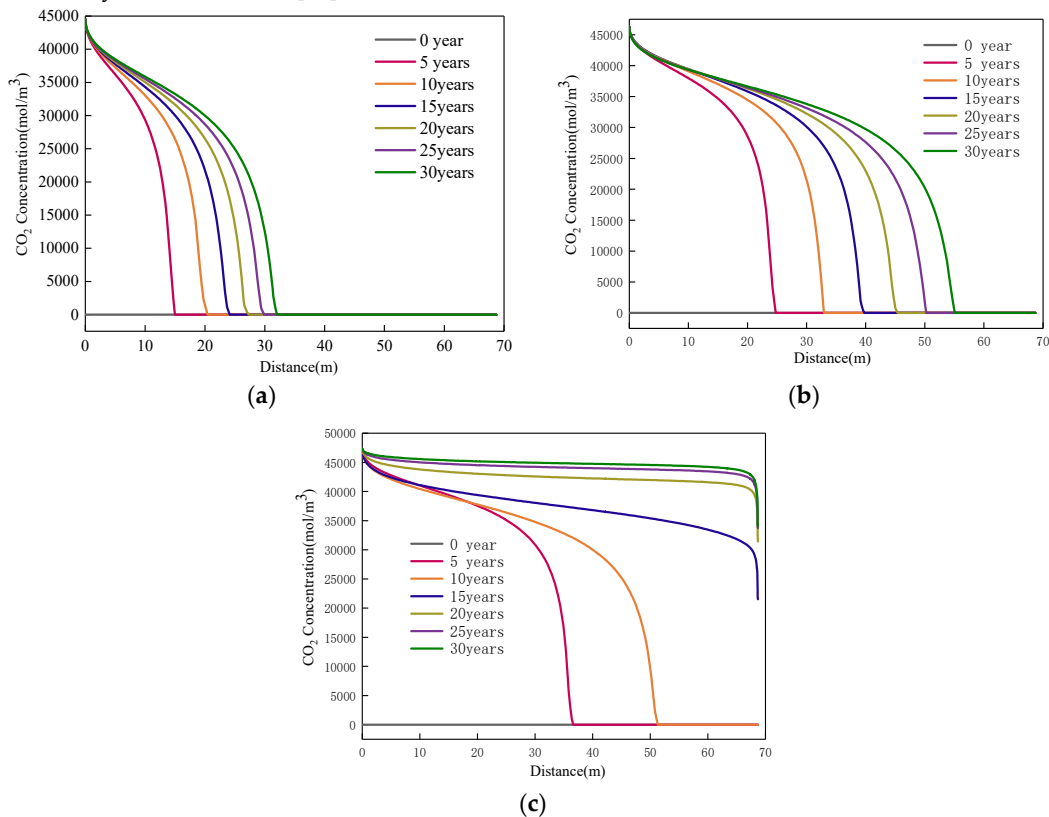
Figure 4 shows the variation of CH<sub>4</sub> molar concentration per unit volume of the diagonal with time. The CH<sub>4</sub> molar concentration increases from the injection well to the displacement front and reaches the maximum value at the latter. The same results can be obtained from the displacement front to the production well because the displacement front is the area with the greatest gas pressure caused by migration. At a certain point along the diagonal of the model, longer displacement times are associated with low CH<sub>4</sub> molar concentrations, and higher injection pressures are associated with faster CH<sub>4</sub> migration and lower molar concentration. After the injected CO<sub>2</sub> pressure reaches 8 MPa for 30 years, the maximum molar concentration of CH<sub>4</sub> in the coal seam is  $1.42 \times 10^3$  (mol/m<sup>3</sup>), and the production rate is 92%.





**Figure 4.** Distribution of diagonal  $\text{CH}_4$  molar concentrations at different injection pressures: (a) injection pressures is 4 MPa; (b) injection pressures is 6 MPa; and (c) injection pressures is 8 MPa.

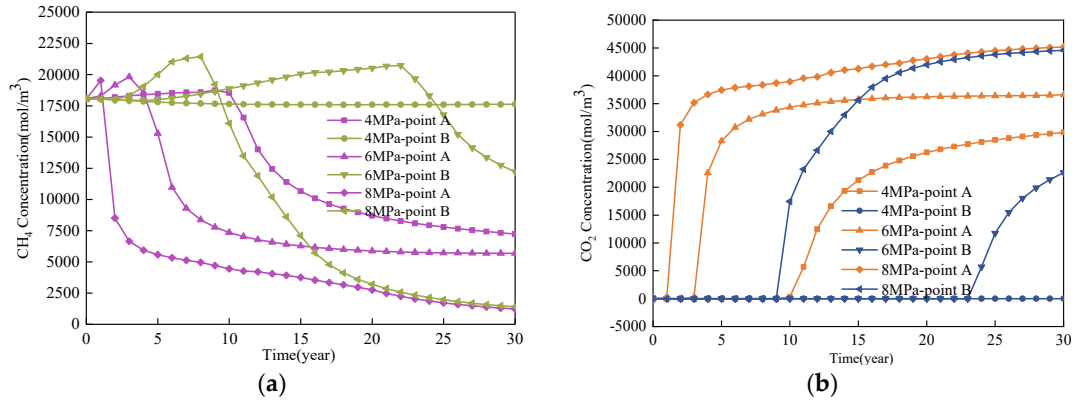
Figure 5 shows the variation of  $\text{CO}_2$  molar concentration per unit volume of the diagonal with time. Shorter distances from the injection well are associated with greater amounts of  $\text{CO}_2$ . There is no  $\text{CO}_2$  from the displacement front to the production well. For a certain point along the diagonal, a longer displacement time and injection pressure lead to more  $\text{CO}_2$  entering the coal seam. The results are basically consistent with [27].



**Figure 5.** Distribution of diagonal  $\text{CO}_2$  molar concentrations at different injection pressures: (a) injection pressures is 4 MPa; (b) injection pressures is 6 MPa; and (c) injection pressures is 8 MPa.

Figure 6a shows the change of  $\text{CH}_4$  molar concentration with time at points A and B simultaneously under different  $\text{CO}_2$  injection pressures. The  $\text{CH}_4$  molar concentration near the injection well A is lower than that far away from the injection well B, and higher injection pressures are associated with higher  $\text{CH}_4$  displacement per unit volume. Figure 6b shows the change of  $\text{CO}_2$  molar concentration with time at points A and B under different injection pressures. Higher pressures

are associated with faster CO<sub>2</sub> migration speeds and higher CO<sub>2</sub> molar concentrations. The displacement occurs first near the injection well. Increasing displacement time shows increased CO<sub>2</sub> molar concentrations in the coal seam.

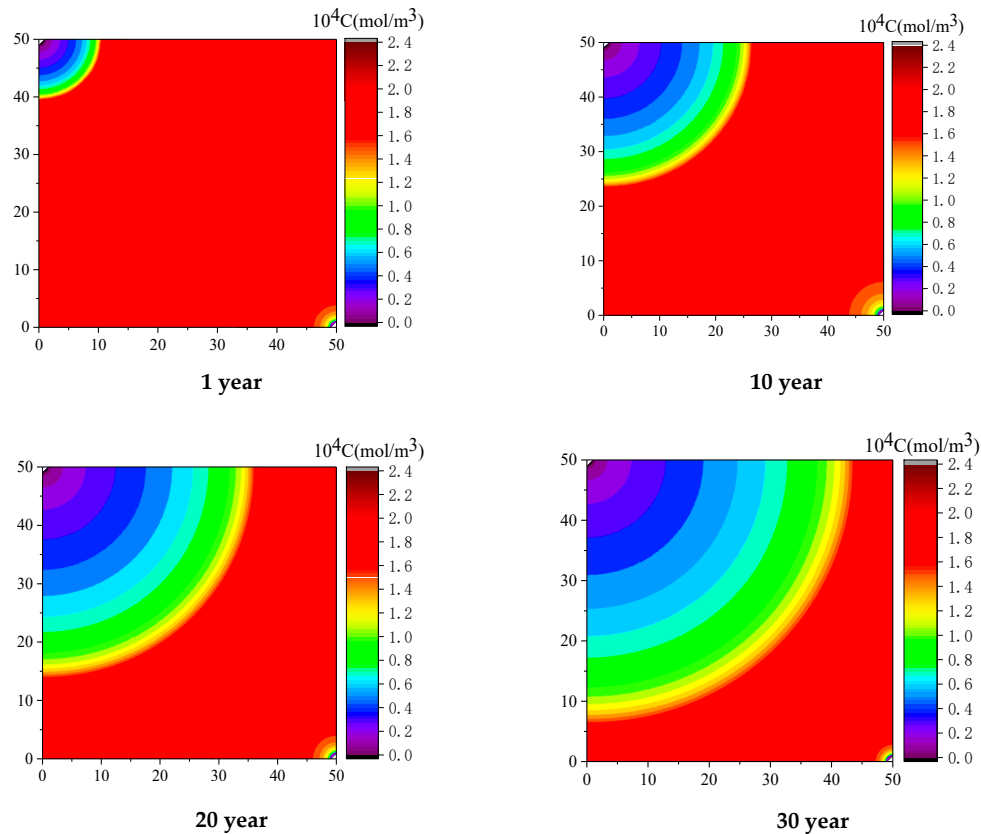


**Figure 6.** CH<sub>4</sub> and CO<sub>2</sub> molar concentrations at monitoring points A and B under different CO<sub>2</sub> injection pressures: (a) CH<sub>4</sub> molar concentration and (b) CO<sub>2</sub> molar concentration.

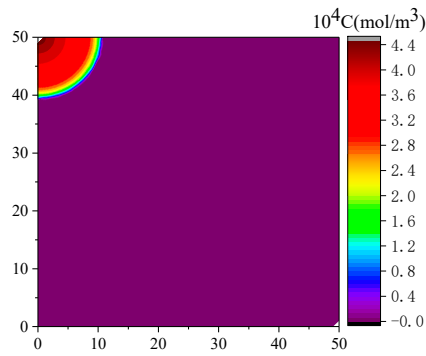
The above analysis shows that high injection pressures are associated with faster gas seepage in the coal seam. CO<sub>2</sub> reserves and CH<sub>4</sub> production both increase with increasing injected CO<sub>2</sub> pressure.

#### 4.2. Effect of Injected CO<sub>2</sub> Temperature on CO<sub>2</sub>-ECBM

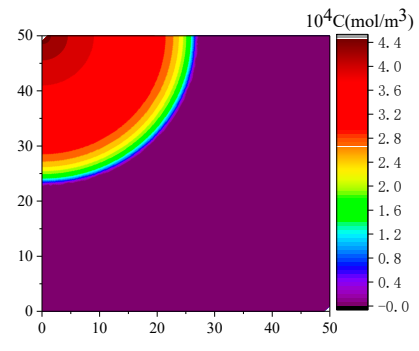
To study the effect of CO<sub>2</sub> injection temperature on CO<sub>2</sub>-ECBM, CO<sub>2</sub> with different temperatures is injected into the coal seam at a fixed pressure, and the distribution rule of CO<sub>2</sub> and CH<sub>4</sub> mole concentrations in CBM over 30 years is investigated. The CO<sub>2</sub> injection temperature is 303, 333, and 363 K, and the pressure of the coal seam is 6 MPa. Figure 7 shows the migration relationship of CH<sub>4</sub> and CO<sub>2</sub> molar concentration with time under different CO<sub>2</sub> injection temperatures.



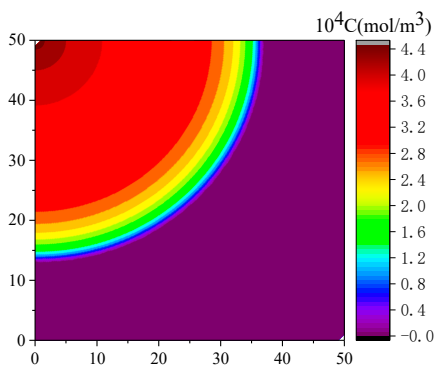
(a)



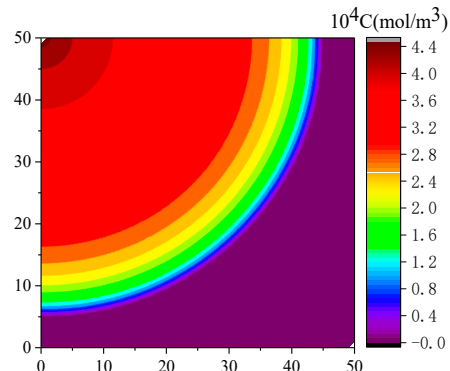
1 year



10 year

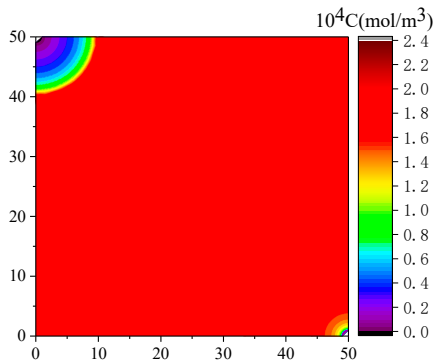


20 year

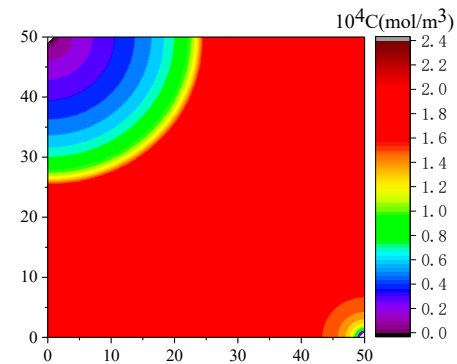


30 year

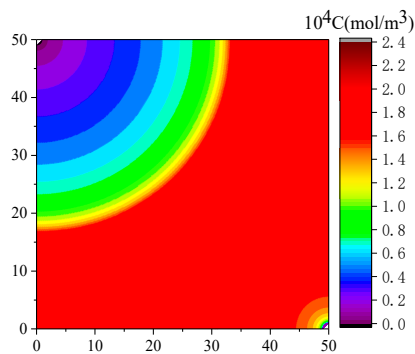
(b)



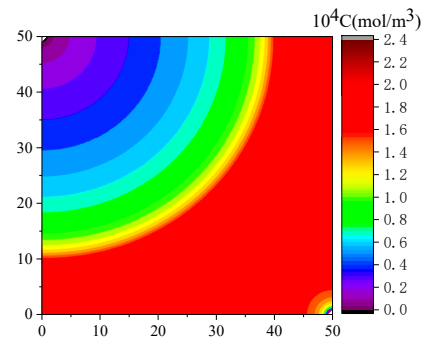
1 year

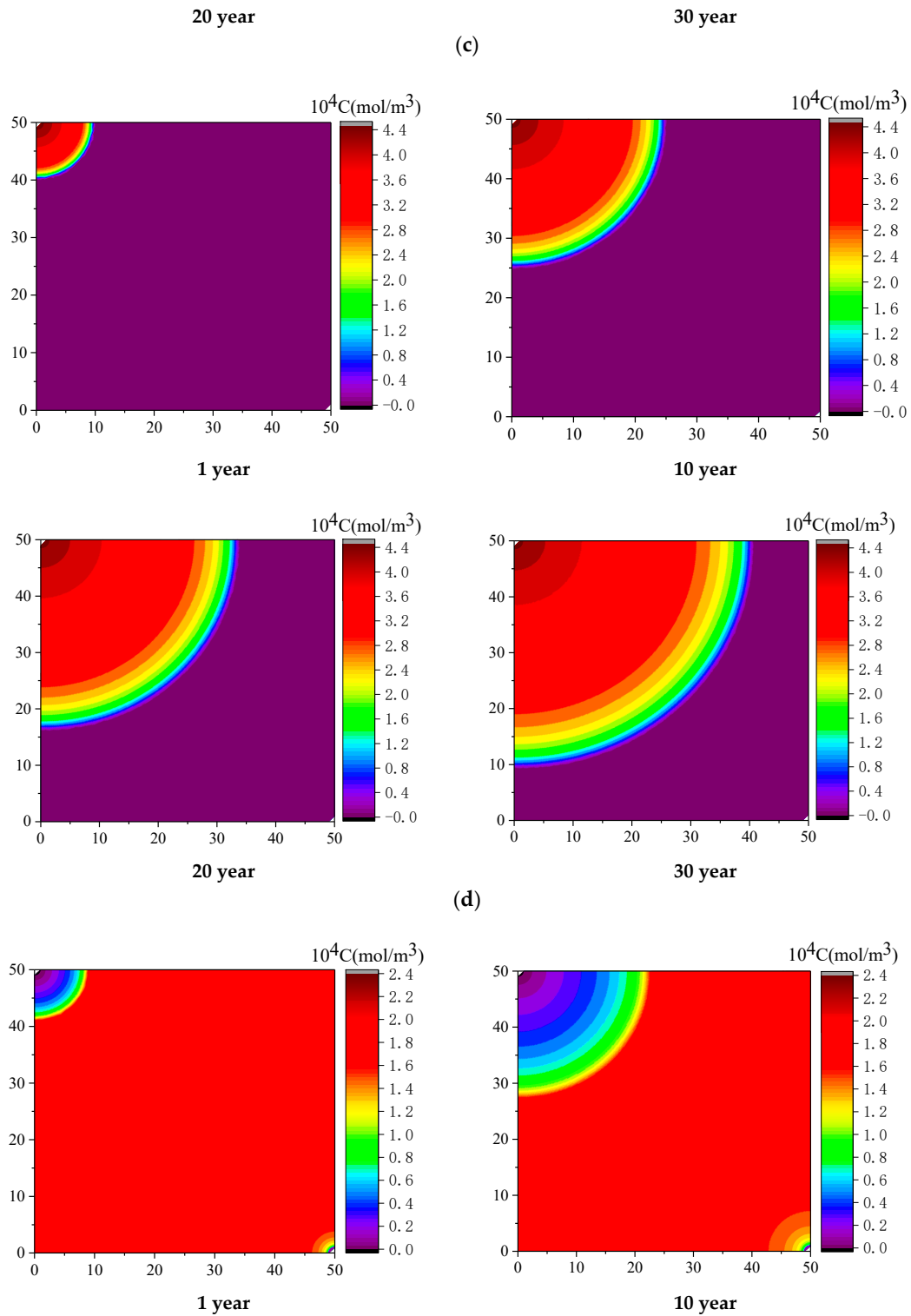


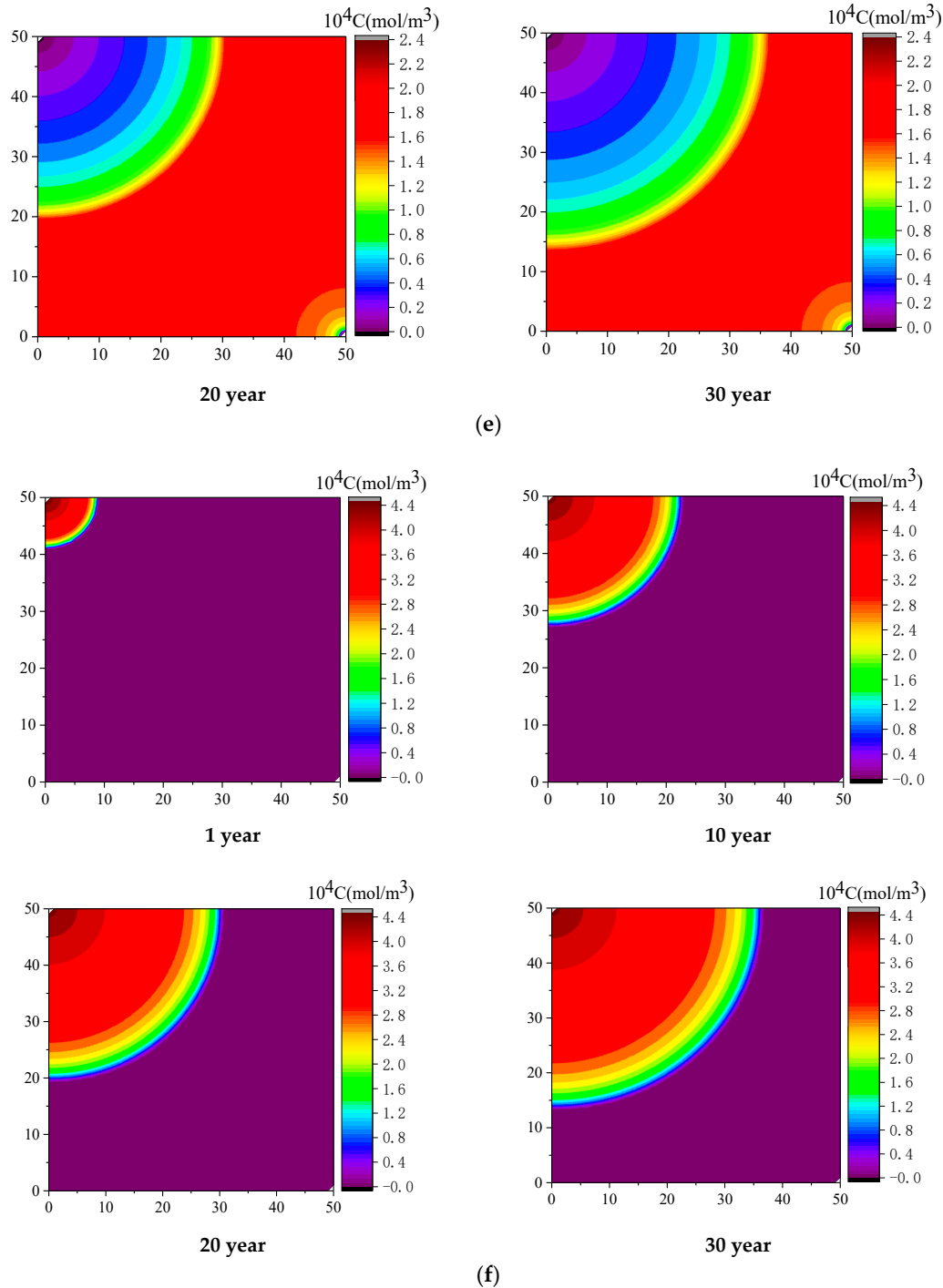
10 year



0





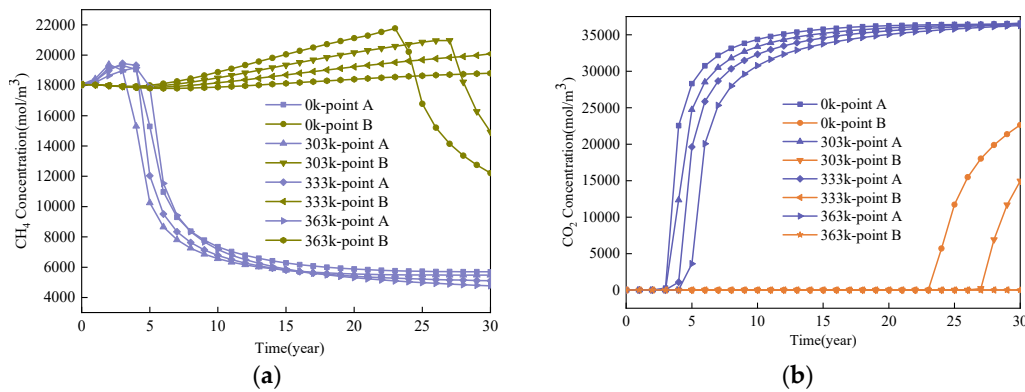


**Figure 7.** Distribution of  $\text{CH}_4$  and  $\text{CO}_2$  molar concentrations under different  $\text{CO}_2$  injection temperatures: (a) distribution of  $\text{CH}_4$  molar concentration at 303 K injection temperatures; (b) distribution of  $\text{CO}_2$  molar concentration at 303 K injection temperatures; (c) distribution of  $\text{CH}_4$  molar concentration at 333 K injection temperatures; (d) distribution of  $\text{CO}_2$  molar concentration at 333 K injection temperatures; (e) distribution of  $\text{CH}_4$  molar concentration at 363 K injection temperatures; and (f) distribution of  $\text{CO}_2$  molar concentration at 363 K injection temperatures.

Figure 7a,c,e shows the molar concentrations of  $\text{CH}_4$  at different temperatures under a  $\text{CO}_2$  injection pressure of 6 MPa. The production of  $\text{CH}_4$  decreases with increasing injection well temperature. After 30 years of heating injection and production, the displacement radius of  $\text{CH}_4$  at

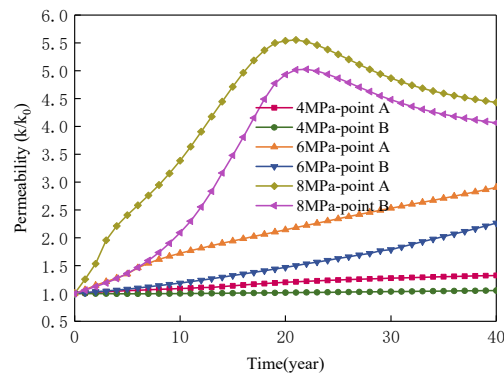
high temperature is smaller than that at low temperature. Figure 7b,d,f are the molar concentrations of CO<sub>2</sub> at different temperatures, also under an CO<sub>2</sub> injection pressure of 6 MPa. After 30 years of heating, the diffusion rate of CO<sub>2</sub> decreases with increasing temperature, which makes the molar concentration of CO<sub>2</sub>-ECBM lower at a high temperature than that at a low temperature under the same pressure.

Figure 8 shows the molar concentrations of CH<sub>4</sub> and CO<sub>2</sub> at points A and B at different temperatures under a CO<sub>2</sub> injection pressure of 6 MPa. The molar concentration of CH<sub>4</sub> at point A at high temperature is lower than that at low temperature, and the law of the molar concentration of CO<sub>2</sub> at point A is the same as that of CH<sub>4</sub>. After 30 years of injection and production, the CH<sub>4</sub> molar concentration at point B is higher at high temperature than at low temperature, while the CO<sub>2</sub> molar concentration is lower than at low temperature. This is because to the coal seam under rising temperature undergoes matrix expansion and decreased permeability. The migration rate of CO<sub>2</sub> decreases owing to the low permeability, which allows it to fully displace CH<sub>4</sub> from the coal seam, but permeability is further reduced at high temperature as a result of CH<sub>4</sub> in the coal seam. The molar concentration of CH<sub>4</sub> production increase is therefore not apparent.



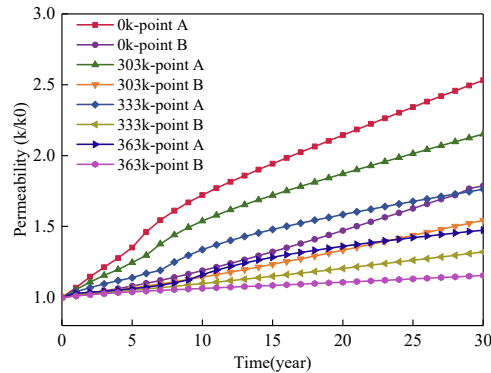
**Figure 8.** CH<sub>4</sub> and CO<sub>2</sub> molar concentrations at monitoring points A and B under different CO<sub>2</sub> injection temperatures: (a) CH<sub>4</sub> molar concentration and (b) CO<sub>2</sub> molar concentration.

Figure 9 shows the changes in permeability ratio of monitoring points A and B when the CO<sub>2</sub> injection pressure is 4, 6, and 8 MPa. The permeability ratio of monitoring points A and B increases over 40 years when the CO<sub>2</sub> injection pressure is 4 and 6 MPa. When the injection pressure is 8 MPa, the permeability ratio initially rises and then declines, and higher injection pressures are associated with higher permeability ratios. This is because the higher injection pressures lead to the full displacement of CH<sub>4</sub> in the coal seam, higher desorption, and higher pore shrinkage of the coal matrix promotes the increase of coal seam permeability. CH<sub>4</sub> desorption decreases in the later stage of injection owing to the large amount of CO<sub>2</sub> absorbed in the coal seam. The swelling effect of the coal matrix is greater than the pore shrinkage and permeability decreases [43–46].



**Figure 9.** Permeability ratios of monitoring points at different injection pressures.

Figure 10 shows the change of permeability ratio of the monitoring points at different temperatures when the CO<sub>2</sub> injection pressure is 6 MPa. Permeability decreases with increasing injection temperature. This is because high temperatures increase the molecular free energy of CO<sub>2</sub> and CH<sub>4</sub>, binary gas molecules become active, CO<sub>2</sub> is not easily adsorbed, and the expansion rate of the coal matrix decreases, leading to reduced permeability.



**Figure 10.** Permeability ratio at the monitoring points at 6 MPa and different injection temperatures.

## 5. Conclusions

We established a thermal–hydraulic–mechanical coupling model of binary gas seepage and diffusion and analyzed the factors influencing conventional production and CO<sub>2</sub>-ECBM. The results are summarized as follows:

- (1) Higher CO<sub>2</sub> injection pressure is associated with higher gas seepage velocities. CO<sub>2</sub> reserves and CH<sub>4</sub> production increase with increasing CO<sub>2</sub> injection pressure. When the injected CO<sub>2</sub> pressure is 8 MPa, the storage capacity of CO<sub>2</sub> is the highest, the radius of effected by CO<sub>2</sub> injection of 5, 10 and 30 years are 31, 44, and 58 m, respectively. After 30 years with injected CO<sub>2</sub> pressure of 4, 6, and 8 MPa, the productivity of CH<sub>4</sub> in the coal seam is 28%, 43%, and 92%, respectively. Therefore, storage of CO<sub>2</sub> and production of CH<sub>4</sub> can be significantly increased by increasing pressure.
- (2) Coal seam temperature has a significant impact on CO<sub>2</sub>-ECBM. Under the same CO<sub>2</sub> injection pressure, CO<sub>2</sub> reserves, CH<sub>4</sub> production, and coal seam permeability all decrease with increasing coal seam temperature. The coal seam matrix shrinks at the beginning due to the desorption of CH<sub>4</sub>, and then it expands due to the adsorption of CO<sub>2</sub> and high temperature. This hinders the seepage and displacement of CO<sub>2</sub>. Therefore, reserve of CO<sub>2</sub>, production of CH<sub>4</sub>, and permeability of coal seam all decrease with increasing coal seam temperature. When the injected CO<sub>2</sub> temperature is 363 K, the storage capacity of CO<sub>2</sub> is the lowest, the radius of effected by CO<sub>2</sub> injection of 10, 20, and 30 years are 22, 30, and 36 m, respectively. After 30 years with injected CO<sub>2</sub> temperature of 303, 333, and 363 K, the productivity of CH<sub>4</sub> in the coal seam is 25%, 22%, and 20%, respectively. The radius of effected by CO<sub>2</sub> injection reduces 10 m when the temperature of CO<sub>2</sub> injection increases from 303 to 363 K. Therefore, high temperatures are not conducive for CO<sub>2</sub> displacement of CH<sub>4</sub>, and the injection temperature should be reduced.

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**Conflicts of Interest:** The authors declare no conflicts of interest.

## Abbreviations

The notations are introduced as follows:

|                    |                                                               |
|--------------------|---------------------------------------------------------------|
| $G$                | The shear modulus, Pa                                         |
| $K$                | The bulk modulus of coal, Pa                                  |
| $E$                | The Young's modulus, Pa                                       |
| $K_S$              | Bulk modulus of coal skeleton                                 |
| $V_m$              | Molar constant of gas                                         |
| $R$                | Universal gas constant, J/mol·K                               |
| $P_i$              | Gas pressure, MPa                                             |
| $T$                | Temperature of the coal, K                                    |
| $\theta_{PY}$      | Stress coefficient caused by gas pressure                     |
| $\theta_{PX}$      | Stress coefficient caused by gas adsorption                   |
| $\theta_T$         | Coefficient of thermal stress                                 |
| $q_g$              | Seepage velocity(m <sup>3</sup> /s)                           |
| $J_i$              | Diffusion component of single phase                           |
| $D_i$              | Diffusion coefficient of single phase, m <sup>2</sup> /s      |
| $m_i$              | Gas mass of each component, kg                                |
| $c_i$              | Molar concentration of a single component, mol/m <sup>3</sup> |
| $k$                | Permeability, m <sup>2</sup>                                  |
| $Q_S$              | Source term, kg/(m <sup>3</sup> ·s)                           |
| $M_i$              | Molar mass of each component, kg/mol                          |
| $dQ_H$             | Heat source for thermal expansion                             |
| $dU$               | The internal energy per unit volume                           |
| $C_V$              | Specific heat at constant volume of coal seam                 |
| $Q_{dis}$          | Differential heat source.                                     |
| $V_{S0}$           | Initial volume of coal skeleton                               |
| $\Delta V_S$       | The coal skeleton volume changes                              |
| $\Delta V_{SP}$    | Bulk expansion and deformation of coal caused by pressure     |
| $\Delta V_{ST}$    | Bulk expansion and deformation of coal caused by temperature  |
| $k_0$              | The initial permeability, m <sup>2</sup>                      |
| $\nu$              | Poisson's ratio                                               |
| $\beta$            | Thermal expansion coefficient, K <sup>-1</sup>                |
| $\delta_{ij}$      | Kronecker function                                            |
| $a_i$              | Langmuir volume constant, m <sup>3</sup> /kg                  |
| $b_i$              | Langmuir pressure constant, Pa <sup>-1</sup>                  |
| $\rho_c$           | Density of coal, kg/m <sup>3</sup>                            |
| $\varepsilon_T$    | Thermal expansion strain                                      |
| $\varepsilon_{PY}$ | Strain caused by gas pressure                                 |
| $\varepsilon_{PX}$ | Gas strain                                                    |
| $\varepsilon_{ij}$ | Train component                                               |
| $u_{ij}$           | Displacement component                                        |
| $\alpha$           | Biot coefficient                                              |
| $\sigma_j$         | Stress tensor( $i, j=1, 2$ )                                  |
| $\mu_i$            | Viscosity coefficient of single phase                         |

|             |                                                          |
|-------------|----------------------------------------------------------|
| $\rho_i$    | Density of a single component, kg/m <sup>3</sup>         |
| $\varphi$   | Porosity                                                 |
| $\rho_a$    | Density of gas at standard conditions, kg/m <sup>3</sup> |
| $\eta$      | Thermal conductivity of coal                             |
| $\varphi_0$ | Initial porosity of coal                                 |
| $\epsilon$  | Volumetric strain of coal                                |
| $d_e$       | Effective diameter of particle                           |

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