Simulations and analysis of hydrate formation during CO$_2$ injection into cold saline aquifers

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Abstract

Geological storage of carbon dioxide (CO$_2$) is perceived to be one of the most promising methods to provide significant reduction in CO$_2$ emissions. During CO$_2$ injected into cold saline aquifers, CO$_2$ hydrate may generate under these temperature and pressure conditions. In the displacement process, the CO$_2$ hydrate formed at the interphase of CO$_2$ and brine could therefore affect the CO$_2$ injection characteristics and the pore structure properties of the aquifers. A multi-phase flow displacement model coupled CO$_2$ hydrate formation process is established in this paper. We use numerical simulations to study characteristics of CO$_2$ hydrate formation during CO$_2$ injection and the displacement process and the permeability evolution of saline aquifers as well. To describe the hydrate formation process, based on the model of hydrate formation kinetics, we employ an improved kinetics model of hydrate formation and derive a thermal energy balance equation for the displacement process, with the thermal effect during the formation of hydrate taken into consideration. The commercial software package COMSOL Multiphysics is used to solve the proposed model numerically. Through numerical simulations, we investigate the CO$_2$ hydrate formation process during the displacement process in the saline aquifer and the impact of hydrate formation on the porosity characteristics and the injection pressure. The results indicate that the formation of CO$_2$ hydrate reduces the porosity of saline aquifers and the velocity of CO$_2$ injected, and the pressure of the wellbore rises gradually, which will impede the CO$_2$ injection and displacement process. During CO$_2$ injection, the process of hydrate formation changes the local temperature and pressure. The remarkable rise in temperature as a result of the exothermic reaction of hydrate formation leads to the reduction of the local hydrate formation rate. In this paper, the simulation and analysis of hydrate formation during CO$_2$ injection into cold saline aquifers will provide references for the optimization of operating conditions and the prediction of CO$_2$ storage in saline aquifers.

Keywords: CO$_2$ hydrate; CO$_2$ injection; Cold saline aquifers; Numerical solution;

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1. Introduction

The concentration of atmospheric CO2 and other greenhouse gases is increasing significantly, due to the increased use of fossil fuels and high-carbon energy sources by human beings. On the premise of maintaining the existing energy structure, carbon capture and storage (CCS) technology is regarded as one of the key technologies to process the generated CO2. Currently, the security issue of CO2 geological storage technology has been considered a top priority of the choice of storage formations, because there will be a serious threat to the ecological environment once CO2 leaks to the surface[1]. During the preparation for an overall long-term CO2 sequestration project, the effective storage technologies with low leakage risk have become an important consideration[2].

When CO2 is injected into the permafrost formations or the brine with high temperature and low pressure, CO2 will combine the water in porous media and form CO2 hydrate. CO2 hydrate is an ice-like crystalline solid material, which is formed by the hydrogen bonding of water molecules wrapping the guest molecules. Water molecules, which are capable of forming hydrogen bonds with each other, assemble together so as to form a cavity wrapping guest molecules. The crystal structure of CO2 hydrate molecular is generally type I (β) structure[3].

In the research of CO2 hydrate formation mechanism, Kvamme et al. apply molecular dynamics, Monte Carlo model, phase field theory and cellular automata model to simulate the nucleation and growth process of CO2 hydrate in seabed sediments. Meanwhile, they analyze how the diffusion-controlled factors affect the hydrate formation process[4]. Tung et al. use the molecular dynamics model to simulate and investigate the growth mechanism of CO2 hydrate and find its growth process is similar with methane’s[5]. Through establishing a dynamic model of the hydration, Kang et al. investigate the rate of hydrate formation. It is shown that with a given temperature and pressure condition, the hydrate formation process will be accelerated if we reduce the temperature or increase the pressure. Furthermore, some hydrate dynamics cocatalysts, such as sodium lauryl sulfate, could significantly accelerate the rate of hydrate formation[6]. The Lattice Boltzmann model is applied in the model by Kang et al., which is established to simulate the growth process of hydrate crystals and to analyze how the relationship between Damkohler number and the saturation of solution affects the growth process of hydrate crystal. When there is a large Damkohler number, the crystal nucleuses are gathered with an increasing saturation of solution. On the contrary, when there is a small Damkohler number, the crystal nucleation process does not change with variational saturation of solution[7].

For the CO2 geological sequestration, Jadhawar et al. use experimental method to simulate that CO2 is injected into depleted oil and gas reservoirs containing methane hydrate[8]. Zatsepina et al. come up with a new approach of CO2 sequestration, which is to sequestrate CO2 with the form of hydrate formed in the reservoirs. They simulate the process of CO2 injection into depleted oil and gas reservoirs, by using the software STARS (Advanced Process and Thermal Reservoir Simulator). It is shown that after the CO2 injection into depleted oil and gas reservoirs, the pressure will increase so that CO2 hydrate could form stably, so as to sequestrate CO2 as solid form largely[9-11]. However, when Vilarrasa et al. study the situation of pressure and temperature variation during the CO2 injection process, they raise the possibility of a phenomenon that, at the area with low temperature and high pressure around the wellbore, CO2 hydrate could form due to the influence of the Joule-Thomson effect in the injection pipeline and the geothermal gradient of the formations[12]. This hydrate formation process could change the temperature and pressure conditions around the wellbore and meanwhile affect the injection process. However, the hydrate formation in the displacement process of CO2 being injected into brine and the influence on CO2 injection process are rarely reported.

The objective of this work is to study the CO2 hydrate formation process during the displacement process of CO2 being injected into low temperature saline aquifer. We investigate the CO2 hydrate formation process during the displacement process and the porosity evolution characteristics of the porous media by numerical simulation. The impact of the porosity evolution for the CO2 injection process is also investigated. In this paper, we describe the physical problem and mathematical models of the CO2 hydrate formation during the displacement process of CO2 being injected into cold saline aquifer. In addition, a function describing the variation of energy in the system of CO2 hydrate formation during the displacement process is derived. Through analyzing the numerical simulation results, we discuss the impact factors on the CO2 hydrate formation process and its influence. Conclusions are given at last.
2. Problem statement

The process of supercritical CO₂ injection into porous saline aquifers involves CO₂ phase (non-wetting phase), brine phase (wetting phase) and the displacement process. When the displacing phase enters the formation from the injection well, the brine will be gradually displaced and meanwhile the two phases interact and transport, as is shown in Fig. 1. With the conduct of the displacement process, CO₂ sweeps the porous media area containing residual brine with low temperature and high pressure so that CO₂ hydrate is formed.

The research in this paper couples a two-phase displacement process and hydrate formation process together, aiming at finding the regular pattern of the impact of the hydrate formation process on the two-phase displacement process and the CO₂ injection process. In this paper, to simplify the above physical model, assumptions are made as follows:

- The saline aquifer is a homogeneous porous medium;
- CO₂ hydrate, brine and CO₂ fluid are the phases in pores individually;
- In the saline aquifer, CO₂ and brine are immiscible phases, and the effect of mutual dissolution is neglected;
- The temperature gradient and hydrostatic pressure gradient are linear distribution. The geothermal gradient is 0.1K/°m and the pressure gradient is 0.01MPa/m;
- At the initial moment, the porous media only contains water without other impurity gases.

3. Governing equations

3.1. Displacement process in porous media

The mass balance equation and momentum equation of the two-phase displacement process in porous media can be expressed as:

\[ \frac{\partial}{\partial t} (\phi \rho_i S_i) + \nabla \cdot (\rho_i \mathbf{u}_i) = \dot{m}_i, \quad i = n, w \]  (1)

\[ \mathbf{u}_i = -\frac{k_{rel,i}}{\mu_i} (\nabla p_i - \rho_i \mathbf{g}), \quad i = n, w \]  (2)

where \( \phi \) is the porosity; \( S_i \) is the saturation of phase \( i \); \( \rho_i \) is the density of phase \( i \); \( \mathbf{u}_i \) is the velocity vector of phase \( i \); \( \kappa \) is the intrinsic permeability tensor; \( k_{rel,i} \) is the relative permeability of phase \( i \); \( \mu_i \) represents the dynamic viscosity of phase \( i \); \( p_i \) denotes the fluid pressure; \( \dot{m}_i \) represents the local mass rate produced by hydrate formation per unit volume. The subscripts ‘n’ and ‘w’ refer to non-wetting phase (CO₂) and wetting phase (brine), respectively.

Since the velocity \( \mathbf{u} \) of CO₂ hydrate phase equals zero, the mass balance equation of CO₂ hydrate could be written as:
\[
\frac{d}{dt}(\phi S_h) = -\dot{m}_h
\]

where \(\dot{m}_h\) represents the local mass rate of hydrate formed per unit volume and the subscript ‘h’ refers to the hydrate phase. The consumption rates of CO\(_2\) and water are related by stoichiometry of the hydrate and related to formation rate of the hydrate by the following:

\[
\dot{m}_h = \dot{m}_n \frac{N_h M_w + M_n}{M_n}
\]

\[
\dot{m}_w = \dot{m}_n \frac{N_h M_w}{M_n}
\]

where \(M_n\) and \(M_w\) are the molecular weight of water and non-wetting phase (in this paper, CO\(_2\)), respectively.

The saturation of each phase meets the following relationships:

\[
S_n + S_w + S_h = 1
\]

For the CO\(_2\)-brine-hydrate three phase displacement-formation system, the van Genuchten-Mualem (VGM) model is applied to describe the relative permeability of each phase[13]:

\[
k_{nw} = S_w^\theta \left( 1 - \left( S_w^{\frac{1}{\theta}} \right)^m \right)^2
\]

\[
k_n = \left( 1 - S_w \right)^\theta \left( 1 - S_w^{\frac{1}{\theta}} \right)^{2m}
\]

In this model, the relationship between effective saturation and capillary pressure is as follows:

\[
S_v = \frac{1}{\left[1 + (\alpha p_c)^\theta \right]^m}
\]

The capillary pressure decides the pressure difference between wetting phase and non-wetting phase. The relationship is expressed as follows:

\[
p_c = p_n - p_w
\]

3.2. Hydrate formation process

The Kim-Bishnoi model could be used to describe the consumption rate function of non-wetting phase (CO\(_2\)) caused by the hydrate formation as follows[14]:

\[
\dot{m}_n = k_f \cdot A_s \cdot f_n \cdot (f_n - f_e)
\]

where \(k_f\) is the formation constant (kg/m\(^2\)-Pa-s); \(A_s\) is the area adjustment factor (dimensionless); \(f_n\) denotes the porous media surface area participating in the reaction; \(f_n\) is the fugacity of CO\(_2\) under the corresponding pressure \(p_n\); \(f_e\) is the fugacity at equilibrium at temperature \(T\) and equilibrium pressure \(p_e\). In this paper, the component pressure is used to displace the component fugacity approximately.

The Kamath (1987) model could express the relationship between hydrate formation pressure \(p_e\) and temperature \(T\) as follows:

\[
p_e = \exp \left( 38.98 - \frac{8533.8}{T} \right)
\]

This equation describes the thermodynamic relationship between hydrate formation pressure \(p_e\) (Pa) and temperature \(T\) (K).

The surface area participating in the reaction is computed by assigning the hydrate saturation uniformly to the interstitial spaces of the porous medium. The original solid grain volume is determined as[15]:

\[
\frac{d}{dt}(\phi S_h) = -\dot{m}_h
\]
where \( r_p \) is the solid grain radius. Then, the number of voids \( (N_v) \) is assumed to be equal to the number of solid grains, and the corresponding void volume \( (V_v) \) is computed from:

\[
V_v = \frac{\phi}{N_v}
\]

where

\[
N_v = \left( \frac{1-\phi}{V_p} \right)
\]

For the reactive area of hydrate formation, the surface area could be written as:

\[
A_i = N_v \left( 4\pi r_p^2 \right)^{2/3}
\]

where the average (effective) radius \( r_p \) of the porous medium grains could be estimated from the Kozeny-Carman equation as follows:

\[
r_p = \left[ \frac{45\kappa_0 (1-\phi)^2}{\phi^3} \right]^{1/2}
\]

where \( \kappa_0 \) is intrinsic permeability of porous media. However, with the formation of hydrate in the porous saline aquifer, the porosity of formations decreases and the permeability changes accordingly. We use the Masuda et al. (1997) model to calculate the change of permeability as follows:

\[
\kappa = \kappa_0 \cdot (1-S_h)^n, \quad n=10,15
\]

### 3.3. Derivation of thermal balance equation

Due to the hydrate formation process accompanied by phase changes, hydrate formation is an exothermic process, which will have impact on the temperature distribution and the process of phase change during hydrate formation. Thus, the system requires a thermal energy balance equation to describe the temperature change of hydrate formation process. The thermal energy balance equation can be expressed as follows:

\[
\frac{\partial}{\partial t} \left[ \rho_i S_i H_i + \rho_i S_i H_s + (1-\phi) \rho_s H_s \right] + \nabla \cdot (\rho_i \mathbf{u}_i H_i) = \nabla \cdot (\lambda \nabla T) + q, \quad i=n, w
\]

where \( H_i \) is the enthalpy of phase \( i; T \) is the system region’s local temperature; \( \lambda \) denotes the heat conductivity of hydrate bearing media; \( q \) denotes the thermal resources of surroundings. The subscript ‘s’ means the sand phase.

From equation (1), (2), (6) and (10), we get:

\[
\frac{\partial}{\partial t} \left[ \phi \rho_i S_i \right] - \nabla \cdot \left( \rho_u \frac{\kappa_m}{\mu_u} \nabla p_u \right) = \dot{m}_u
\]

\[
\frac{\partial}{\partial t} \left[ \phi \rho_u (1-S_u-S_s) \right] - \nabla \cdot \left( \rho_u \frac{\kappa_m}{\mu_u} (\nabla p_u - \nabla p_s) \right) = \dot{m}_w
\]

The relationship of components’ latent heat during hydrate formation is defined as follows:

\[
\dot{m}_u H_u + \dot{m}_w H_w = \dot{m}_h \Delta H_f
\]

where \( \Delta H_f \) is defined as the enthalpy change of hydrate formation.

Inserting equation (3), (20), (21) and (22) to equation (19), we get:

\[
(1-\phi) \frac{\partial}{\partial t} (\rho_i H_i) + \phi \left( \rho_s S_u \frac{\partial H_u}{\partial t} + \rho_s S_w \frac{\partial H_w}{\partial t} + \rho_s S_h \frac{\partial H_h}{\partial t} \right) + \rho_u \mathbf{u}_u \cdot \nabla H_u + \rho_u \mathbf{u}_w \cdot \nabla H_w = \nabla \cdot (\lambda \nabla T) + \dot{m}_h \Delta H_f
\]
Note that
\[ \frac{dH}{dT} = \frac{\partial H}{\partial T} + \frac{\partial H}{\partial p} \frac{dp}{dT} = C_{p_i} \frac{dT}{dT} + \sigma_i \frac{dp_i}{dT}, \quad i=n,w,h \] (24)
where the term \( \sigma_i \) represents the Joule-Thomson (J-T) coefficient of phase \( i \) in the system due to the process we investigate involving J-T effect. For the sand phase,
\[ \frac{dH}{dT} = \frac{\partial H}{\partial T} \frac{dT}{dT} = C_{p_s} \frac{dT}{dT} \] (25)

For the thermal energy balance equation of the system, we neglect the impact of pressure on the enthalpy of water phase and hydrate phase, as well as the inter-phase heat transfer process. Inserting equation (24) and (25) to equation (23), we get the final form of thermal energy balance equation as follows:
\[ \begin{align*}
\phi \rho_n S_n C_{pm} + \phi \rho_w S_w C_{pw} + \phi \rho_s S_s C_{ps} + (1 - \phi) \rho_s C_{ps} & \left( \frac{\partial T}{\partial t} + \left( \rho_s u_s C_{ps} + \rho_s u_s C_{ps} \right) \cdot \nabla T \right) \\
- \phi \rho_n S_n \sigma_n \frac{\partial p_n}{\partial t} - \rho_w \sigma_n \cdot \nabla p_n & = \nabla \cdot \left( \lambda \nabla T \right) + \hat{m}_n \Delta H_f
\end{align*} \] (26)

3.4. Initial and boundary conditions

The initial conditions and the boundary conditions for each phase are as follows:
The initial conditions of non-wetting phase are:
\[ p_n \big|_{t=0} = \rho_n g (H - z) + p_d \] (27)
The initial conditions of wetting phase are:
\[ p_w \big|_{t=0} = \rho_n g (H - z) \] (28)
The boundary conditions of non-wetting phase are:
\[ u_n \big|_{r=R} = u_0 \] (29)
\[ u_n \big|_{z=0} = 0, \quad u_n \big|_{z=H} = 0 \] (30)
\[ p_n \big|_{r=R} = \rho_n g (H - z) + p_d \] (31)
The boundary conditions of wetting phase are:
\[ u_w \big|_{z=0} = 0, \quad u_w \big|_{z=H} = 0 \] (32)
\[ p_w \big|_{r=R} = \rho_n g (H - z) \] (33)

4. Results and discussion

4.1. Numerical simulation

The major objectives of this paper are to investigate the CO2 hydrate formation during CO2 injection and displacement process and the evolution characteristics of the CO2 injection process. A computational domain with a 30m height and 100m radius is established by applying a cylindrical coordinate, describing that CO2 is injected into saline aquifers through vertical injection wells associated with a two-phase displacement process. CO2 is injected into the formation from the wellbore is placing the brine phase in the porous media of the saline aquifer. CO2 contacts the residual water in the pores and forms hydrate at some areas of the region. A commercial software COMSOL Multiphysics 4.0 is applied in this paper to numerically solve the mathematical model. The relevant parameters used for the simulations are given in Table 1. The injection durations simulated were up to 150 days. The temperature and pressure conditions of injection process are: \( T = 278K, \quad P = 10MPa \).
Table 1. Properties and operational conditions in the simulations

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Aquifer</strong></td>
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</tr>
<tr>
<td>Aquifer thickness $H$ (m)</td>
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</tr>
<tr>
<td>Intrinsic permeability $K_0$ (m$^2$)</td>
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<tr>
<td>Porosity $\phi$</td>
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</tr>
<tr>
<td><strong>CO$_2$</strong></td>
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</tr>
<tr>
<td>Viscosity $\mu_a$ (Pa·s)</td>
<td>$3.95 \times 10^{-5}$</td>
</tr>
<tr>
<td><strong>Brine</strong></td>
<td></td>
</tr>
<tr>
<td>Density $\rho_w$ (kg·m$^{-3}$)</td>
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</tr>
<tr>
<td>Viscosity $\mu_w$ (Pa·s)</td>
<td>$2.54 \times 10^{-4}$</td>
</tr>
<tr>
<td>Gravitational acceleration $g$ (m·s$^{-2}$)</td>
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</tr>
<tr>
<td><strong>Hydrate</strong></td>
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<tr>
<td>Hydrate number $N_h$</td>
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</tr>
<tr>
<td>Formation kinetics constant $k_f$ (kg·m$^{-2}$·Pa$^{-1}$·s$^{-1}$)</td>
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</tr>
<tr>
<td>Density $\rho_h$ (kg·m$^{-3}$)</td>
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</tr>
<tr>
<td>Heat capacity $C_{ph}$ (J·kg$^{-1}$·K$^{-1}$)</td>
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</tr>
<tr>
<td><strong>Operation parameters</strong></td>
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<tr>
<td>Injection rate $u_0$ (m$^3$·day$^{-1}$)</td>
<td>1600</td>
</tr>
</tbody>
</table>

4.2. **Hydrate formation during displacement process**

Fig. 2 shows us how the saturation of CO$_2$ hydrate phase changes with the change of time and space during 150 days.

![CO$_2$ hydrate saturation changes over time](image)
As is shown in Fig. 2, after CO₂ is injected into the brine, the saturation of CO₂ hydrate decreases along the radius of entire calculation region. However, the saturation of CO₂ hydrate increases gradually as time goes on, which means that CO₂ hydrate continues to form and accumulate over time. There is a larger CO₂ hydrate saturation near the injection wellbore, and the amount of CO₂ hydrate formation declines along with the radius. With the front of CO₂ fluid phase progressing forward continually, the front of formed CO₂ hydrate moves forward over time as well. The amount of CO₂ hydrate accumulation increases continually at the injection wellbore.

![Fig. 3. Relationship between the system temperature and the local hydrate formation rate](image)

Fig. 3 shows the temperature distribution of the system at the time of the 150th day. As is seen from the figure, the CO₂ hydrate formation reaction is an exothermic reaction. In the process of temperature rising, the rate of hydrate formation decreases. When the temperature reaches the peak value point and then begins to decrease, the rate of hydrate formation reaches the minimum value point and then begins to increase correspondingly.

![Fig. 4. Distribution of CO₂ velocity at the 150th day](image)

Fig. 4 shows the velocity distribution of CO₂ fluid and pressure distribution curve at the moment of the 150 day. As is shown in the figure, the CO₂ velocity decreases sharply near the injection wellbore with a sharp rise of pressure at the same time. After CO₂ is injected into saline aquifers, the porosity of the brine is significantly reduced due to a large amount of formed hydrate; as a result the velocity decreases dramatically and the pressure increases.

![Fig. 5. Distribution of CO₂ hydrate saturation and porosity of saline aquifers at the time of the 150th day](image)

Fig. 5 shows the distribution of CO₂ hydrate saturation and porosity of saline aquifers at the time of the 150th day. As is shown in Fig. 5, CO₂ hydrate is mostly found near the injection wellbore and it declines along with the radius. Therefore, the porosity of the saline aquifer is minimal near the injection wellbore and gradually increases with the reduction of the amount of the hydrate accumulation.
4.3. Evolution of temperature and pressure when CO\textsubscript{2} hydrate forms during displacement process

Fig. 6 shows the relationship between the local temperature and time of the system. Since the CO\textsubscript{2} hydrate formation reaction is an exothermic process, the rising temperature will impede the hydrate formation. In this paper, the computational temperature conditions at the injection wellbore are the constant temperature of formations. Therefore, with the increasing amount of hydrate formation, the local temperature increases, which affects the rate of CO\textsubscript{2} hydrate formation. According to the computational conditions used in this paper, the temperature of the system increased by about 4°C within 150 days. In addition, with the continuous CO\textsubscript{2} injection and the ongoing displacement process, the local temperature peaks move forward along with the direction of displacement process.

Fig. 7 shows the simulation results of the system pressure distribution along with the radius at different time. As is shown in the figure, the pressure distribution is downward along with the radius, but the pressure at the injection wellbore is increasing, which is caused by the decreasing porosity due to the continuous CO\textsubscript{2} formation. The CO\textsubscript{2} fluid velocity decreases and the pressure rises near the injection wellbore, which increases the difficulty of CO\textsubscript{2} injection process.
5. Conclusions

A multi-phase flow displacement model coupled with CO$_2$ hydrate formation process in porous media is established in this paper. To describe the hydrate formation process, based on the model of hydrate formation kinetics, we employ an improved kinetics model of hydrate formation and derive a thermal energy balance equation for the displacement process, with the thermal effect during the formation of hydrate taken into consideration. The commercial software package COMSOL Multiphysics is used to solve the proposed model numerically. Through numerical simulations, we investigate the CO$_2$ hydrate formation process during the displacement process in the saline aquifer and the impact of hydrate formation on the porosity characteristics and the injection pressure. The results indicate that the formation of CO$_2$ hydrate reduces the porosity of saline aquifers and the velocity of CO$_2$ injected, and the pressure of the wellbore rises gradually, which will impede the CO$_2$ injection and displacement process. During CO$_2$ injection, the process of hydrate formation changes the local temperature and pressure. The remarkable rise in temperature as a result of the exothermic reaction of hydrate formation leads to the reduction of the local hydrate formation rate. In this paper, the simulation and analysis of hydrate formation during CO$_2$ injection into cold saline aquifers will provide references for the optimization of operating conditions and the prediction of CO$_2$ storage in saline aquifers.

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