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Review

Review on the Mechanism of CO_2 Storage and Enhanced Gas Recovery in Carbonate Sour Gas Reservoir

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Abstract: Carbonate gas reservoirs in the Sichuan Basin have many complex characteristics, such as wide distribution, strong heterogeneity, high temperature, high pressure, high H_2S and CO_2 content and an active edge or bottom water. In the late stage of exploitation of carbonate sour gas reservoirs, the underground depleted reservoirs can provide a broad and favorable space for CO_2 storage. If CO_2 is injected into the depleted carbonate sour reservoirs for storage, it will help to achieve the goal of carbon neutrality, and the CO_2 stored underground can perform as "cushion gas" to prevent the advance of edge or bottom water, to achieve the purpose of enhanced natural gas recovery. Injecting CO_2 into low permeability reservoirs for oil displacement has become an important means to enhance oil recovery (EOR). However, the mechanism of EOR by injecting CO_2 into carbonate sour gas reservoirs is not clear and the related fundamental research and field application technology are still in the exploration stage. This paper reviews the main scientific and technical perspectives in the process of injecting CO_2 into carbonate sour gas reservoirs for storage and enhancing gas recovery.

Keywords: sour gas reservoir; CO_2 storage; enhanced gas recovery; phase; water–rock reaction

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1. Introduction

Energy is the crucial to the economy and the quality of people's life. The oil and gas industry has a new mission of the development of a high-quality "carbon neutral" and "energy revolution" industry. China is the world's second largest oil consumer and the third largest natural gas consumer, and its oil and gas external dependence has reached 72.2% and 46%, respectively; the oil and gas security situation is exceptionally serious. In addition, carbon peaking and carbon neutrality are major strategic initiatives for China. Thus, reducing carbon emission while sustaining oil and gas production is a critical issue.

China's Sichuan Basin is rich in natural gas resources, and contains 23% and 26% of the country's conventional and unconventional gas resources, respectively. By increasing development efforts, the Sichuan Basin will account for one-third of total domestic natural gas production in the future. Among them, tight gas and shale gas resources are abundant, with remaining recoverable resources of over 7 trillion cubic meters. Carbonate sour gas reservoirs are highly productive, and the depleted carbonate sour gas reservoirs are favorable sites for CO_2 storage. The underground storage capacity of CO_2 in the depleted carbonate sour gas reservoirs can be as high as 7 billion tons, which can virtually solve the problem of carbon neutrality in Sichuan for decades to come.

The following major scientific issues and technical problems exist in the exploitation of various types of gas reservoirs including sour gas reservoirs, tight gas reservoirs and shale gas reservoirs in the Sichuan Basin for CO_2 sequestration and enhanced gas recovery.

(1) The phase state characteristics of CO_2 multi-component mixed system and natural gas multi-component coexistence systems are complex.
(2) The law of percolation, migration, and storage of CO_2 mixed system in reservoir is still unclear.

(3) The mechanism of underground CO_2 storage to form "cushion gas" to prevent edge/bottom water intrusion and CO_2 displacement to drive natural gas is unclear.
(4) The evaluation system of CO_2 storage potential and EOR potential of heterogeneous gas reservoirs with different geological characteristics has not been formed, so it is difficult to carry out large-scale field test and application.

The mechanism of CO_2 sequestration and EOR in the depleted carbonate sour gas reservoirs in Sichuan Basin is still unclear, and relevant fundamental research and field application technologies are still in the exploratory stage. Existing theoretical and experimental studies cannot reveal the mechanism of CO_2 sequestration and EOR in depleted carbonate sour gas reservoirs in Sichuan Basin. It is necessary to use the combination of physical simulation and numerical simulation to carry out related research.

2. Review on the Mechanism of CO_2 Storage and Enhanced Gas Recovery

2.1. Current State of Thermodynamics Research on Solubility and Phase Equilibrium of Supercritical CO_2 Multi-Component Coexistence Systems

Scholars around the world have conducted a large amount of research on the solubility of CO_2 in saline system. Diamond and Akinfiev [1] calculated the solubility of CO_2 in pure water within the pressure range of 0.1~100 MPa and the temperature range of −1.5~100 °C. Tong [2] used the bubble point method to measure the solubility of CO_2 in $CaCl_2$ solution, $MgCl_2$ solution, and synthetic brackish water under the pressure of 40 MPa and the temperature of 423 K. The experiment showed that temperature and pressure basically did not affect the CO_2 solubility in $CaCl_2$ and $MgCl_2$ solutions with the same concentration. Poriter and Rochelle [3] conducted a phase study in CO_2-Utsira oilfield when the temperature range was 273–573 K and the pressure range was 0.1–30 MPa, and the results were applied in the world's largest CO_2 Underground Saltwater Storage Project (Spleipher). Under the condition of temperature range of 308~328 K and pressure of 16 MPa, Liu [4] studied the phase state changes of CO_2 in NaCl, KCl, and $CaCl_2$, and in the mixed solution of NaCl, KCl, and $CaCl_2$ by using the method of water bath heating. The results showed that the solubility of CO_2 in KCl solution is higher. Under the temperature range of 0~80 °C and the pressure range of 8~12 MPa, Hu [5] measured the solubility of CO_2 in the underground water of Shanxi Formation in the Ordos Basin through experiments. Wang [6] sampled and analyzed the brine of several reservoirs in Ordos Basin and completed the CO_2 solubility experiment under the conditions of temperature range of 40~70 °C and pressure range of 8~11 MPa. Jin [7] used Raman spectroscopy to test the solubility of CO_2 in salt solution at the burial depth of 800–2800 m and the salinity of 8.25–99.0 g/L. The experiment results showed that the solubility of CO_2 decreases with the increase in the salinity, and first decreases, then increases with the increase in the burial depth. The critical data of common supercritical fluids in carbonate sour gas reservoirs are shown in Table 1

Table 1. Critical data table of common supercritical fluids in carbonate sour gas reservoirs.

Substance	Critical Temperature/°C	Critical Pressure/MPa
CH_4	−83.00	4.60
C_2H_6	32.40	4.89
CO_2	31.06	7.39
H_2S	100.20	8.94

The phase states of supercritical CO_2-conventional natural gas system have been extensively studied around the world [8–10]. However, due to the strong corrosive and extremely toxic nature of H_2S, the safety requirements in the laboratory greatly limit the phase experiments of the multi-component coexistence systems of supercritical CO_2-brine and sour gas. Therefore, most scholars study the thermodynamic model of phase equilibrium in the CO_2-sour gas system.

Carroll [11,12] analyzed the phase equilibrium prediction model and verified the model when the components contained a mixture of CO_2 and H_2S. Dubessy [13] established a phase equilibrium model suitable for CO_2-NaCl salt solution and H_2S-NaCl salt solution, which has good applicability when the temperature is above 100 °C. Based on PR equation of state, Mireault [14] conducted relevant research and sensitivity analysis on acid gas containing CO_2 and H_2S in wellbore. Guo [15,16] improved the Chrastil model, fitted the coefficients of the new model, and constructed a new sulfur solubility prediction model containing three parameters (temperature, pressure, and gas density). Zhao and Lvov [17] brought up a phase equilibrium model of CO_2-methane-water system by improving the binary interaction model and modifying the PR state equation, and the model was verified with experimental data. Ababneh and Al-Muhtaseb [18] established a phase equilibrium thermodynamic model of CH_4-CO_2-H_2S ternary system, which can better predict the phase state changes of multi-component gas during the development of sour gas reservoirs. Wang and Tang [19] established a wellbore flow and phase distribution model considering the reinjection condition of CO_2-H_2S system. Zhang [20] conducted a phase simulation study of the coexistence systems of natural gas and water containing CO_2 and H_2S, and conducted the sensitivity analysis of pressure, temperature, water content, and other factors. In the experiment of CO_2-H_2S coexistence systems, Theveneau [21] and Souza [22], have conducted phase equilibrium experiment of CO_2-H_2S-CH_4 system. Zhang [23] conducted a phase equilibrium experiment of CO_2-H_2S-crude oil system. The experiment results have a certain guiding significance for the study of CO_2 storage in carbonate sour gas reservoirs, but the experiment temperature and pressure conditions and the content of components are still different from the actual condition in the reservoirs.

In conclusion, there have been a large number of studies on the dissolution of CO_2 in brine system. Scholars from different countries have studied the solubility of CO_2 in different brine systems and under different temperature and pressure, which provides a large amount of foundation for subsequent studies. However, due to the highly toxic and corrosive nature of H_2S, most of the research are phase equilibrium thermodynamic studies. Experimental studies on the coexistence of supercritical CO_2, brine and sour gas are still at a standstill. Thus, it is a scientific and technical problem that must be solved to study the phase states of the multi-component coexistence systems of supercritical CO_2, brine and sour gas. Phase experiments and thermodynamic models of phase equilibrium for similar systems are still scarce.

2.2. Supercritical CO_2-Water–Rock Reaction Experiment and Coupled Kinetic Model

The interaction between supercritical CO_2 and rocks can cause mineral dissolution, precipitation, hydrolysis, diffusion, and other physicochemical phenomena. Usually, the techniques, such as cast sheet identification, environmental scanning electron microscopy, X-ray diffraction, NMR, constant velocity mercury injection, and CT scanning, are used to study the physical parameters of rocks and their changes, pore microstructure changes, and analyze the mineral composition and wettability of rocks [24,25]. Shao [26] used atomic force microscopy (AMF) and found that the reaction between CO_2-water-rock would cause dissolution of mica minerals, and, at the same time, nanoscale sediments were generated near the dissolution hole. Guo [27] used X-ray diffraction instrument, multi-function ion chromatograph, ultra-low permeability gas permeability measuring instrument and environmental scanning electron microscope to conduct experimental research. Their results showed that the CO_2-H_2S-water–rock reaction would lead to the dissolution of some rock minerals, thus increasing the porosity and permeability of the reservoir. Tang [28] analyzed the dynamic mechanism of brine and rock mineral properties by the interaction of CO_2-brine-rock by scanning electron microscopy, energy spectrometer, X-ray diffractometer and flame atomic absorption spectrometry. The results showed that the CO_2–brine–rock reaction caused more serious damage to the gas reservoir than the aquifer. At the same time, the reservoirs with poor physical properties are more damaged than those with good physical properties. For carbonate sour gas reservoirs, the destruction of reservoir physical properties is more serious due to liquid sulfur adsorption and sulfur fixation deposition [29]. Liu [30] used PHREEQC software to simulate the effect of fluid temperature, pH, ion content, CO_2 partial pressure, and other factors on mineral dissolution in the process of water–rock reaction. Dong [31] applied TOGUHREACT to simulate the changes of formation pressure, fluid pH, ion content, mineral dissolution, precipitation, reservoir physical properties, and CO_2 storage, and studied the influence of CO_2–water–rock interaction on reservoir properties. Pearce [32] experiment found that in sandstone rich in chlorite, when CO_2 reacts with low concentration SO_2, rock porosity will not increase, but there is particle movement, which damages permeability. Ahmat [33] In order to better understand CO_2–water–rock interaction, Different mineral compositions and fluid salinity were numerically simulated and experimentally analyzed. The effects of different mineral compositions in different stages of CO_2–water–rock interaction and the differences of products were analyzed. Wang [34] studied the CO_2–water–rock interaction system in carbonate rocks containing calcite and dolomite minerals by using the PHREEQC geochemical package through dynamic and static experiments, and carbon dioxide mainly exists on the surface of minerals through carbonate ions. Tang [35] and Huang [36] conducted an experimental study on the changes of macroscopic rock mechanical properties under different lithologies, different soaking methods and different flow rates of aqueous solution. Result showed that the water–rock reaction reduced the compressive strength and elastic modulus of rock. The change of rock mechanical properties due to the water–rock reaction is related to the PH value of water, hydrolysis, dissolution, ion exchange, temperature, pressure, and other factors in the reaction. Nguyen [37–39] experimentally studied the changes of macroscopic mechanical properties and microscopic morphological characteristics of sandstone samples caused by the water–rock reaction at different temperatures, pressures, PH values, lithologies, fluid components, injection rates, and soaking methods. Due to the influence of chemical alteration, hydration, hydrolysis, dissolution, and ion exchange, the elasticity and strength of rocks are compromised. Feng [40] analyzed the influences of water chemical corrosion on rock triaxial compressive strength, uniaxial compressive strength, shear strength, tensile strength, and cracking characteristics. Lu [41] conducted water–rock reaction experiments in fractured sandstone and analyzed the influence of fine microstructure and structural chemical corrosion on water–rock reaction. Smith [42] conducted field tests combined with hydrogeological surveys, and the results showed that the self-occurrence of carbonate reservoirs and the deposition of anhydrite layer during water–rock reaction had a positive effect on the storage of CO_2–H_2S acid gas. In addition, even if the anhydrite layer was fractured, the dissolution and mineralization of the overlying aquifer will also slow the movement of acid gas and prevent it from migrating to the surface. Guo [43] conducted experiments on water–rock reaction in sour gas reservoirs, which established the dynamic model and numerical simulation model of acid gas–water–rock reaction. The study also described the mechanism of water–rock reaction, and the behavior of reservoir physical properties, sulfur deposition, and their combined effects on reservoir physical properties. Acidic gas has an evident corrosion effect on marine carbonate rocks under high temperature and high-pressure environment, which increases the permeability and porosity of reservoir core gradually, and H_2S gas has a better dissolution effect on reservoir rocks than CO_2 gas. The high temperature and high-pressure water–rock reaction device is shown in Figure 1.

In terms of the coupling dynamics of water–rock reaction, Tan [44] established the coupling dynamic model of reaction-transport-mechanics (RTM) of the stratiform-fluid-metallogenic system and proposed the application of the incremental stress rheology formula, which further deepened the understanding of the coupling dynamics of water–rock reaction. Li [45], based on the non-Darcy percolation theory, analyzed the coupling mechanism of resistance and dynamic force in the process of oil and gas migration and the characteristics of oil and gas accumulation under the action of this mechanism. Wang [46] established a near-field dynamic model of rock mass coupled with thermal-water-mechanical-chemical coupling based on the non-local continuum mechanics theory. The coupling kinetics model of reaction-Transport-mechanics (RTM) involves many fields, including chemistry, mechanics and chemistry, percolation mechanics, hydrogeology, and thermodynamics. They are irreversible processes and generally have nonlinear dynamics mechanism. Therefore, the introduction of complexity theory provides conditions for the establishment of reaction-transport-mechanics model and the exploration of the space-time evolution process of complex dynamics of water–rock reaction. Table 2 shows the reaction-ion relationship between water and rock in carbonate sour gas reservoirs.

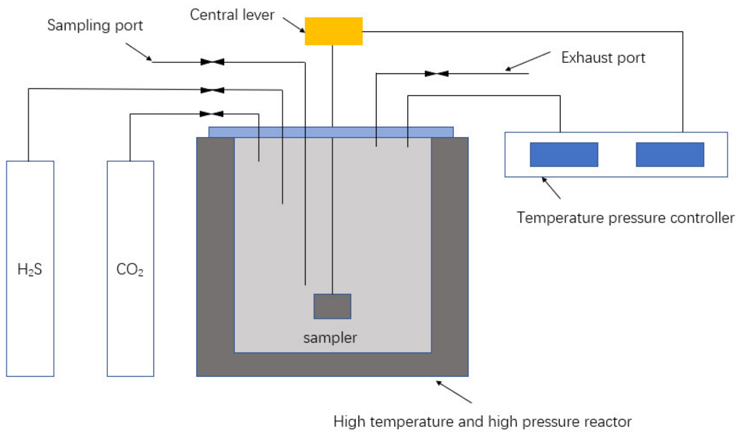

Figure 1. High temperature and high-pressure water–rock reaction device.

Table 2. Microscopic ion reaction process of water–rock reaction in carbonate sour gas reservoirs.

Ionic Reaction Formula
$H_2S = H^+ + HS^-$
$CO_2 + H_2O = H^+ + HCO_3^-$
$Mg{\bullet}CaCO_3 + 2H^+ = Ca^{2+} + Mg^{2+} + CO_2 + H_2O$
$Ca^{2+} + CO_2 + H_2O = CaCO_3{\downarrow} + 2H^+$
$HS^- + Fe^{2+} = FeS{\downarrow} + 2H^+$

In summary, many scholars have studied the products and influencing factors of water–rock reaction, analyzed the influence of water–rock reaction products on rock mechanical properties and CO_2 storage, and combined water–rock reaction with other disciplines, creating conditions for the complex dynamic space-time evolution process of water–rock reaction. However, there are few reports on experiments and theories of physical and chemical reactions of supercritical CO_2-brine-rock in high-temperature and high-pressure sour gas reservoirs. Therefore, more comprehensive experiments and theoretical studies need to be conducted.

2.3. *Recent Research on Diffusion, Transport, and Flow Law of Supercritical CO_2*

The measurement methods of CO_2 diffusion coefficient mainly include direct method and indirect method. Indirect methods include nuclear magnetic resonance (NMR) [47,48], PVT simulation method [49], and dynamic drop method [50]. The direct method is based on Fick's law and combined with the gas equation of state and the continuity equation. According to the designed experimental equipment and process, the corresponding initial conditions and boundary conditions are given, we can obtain the mathematical model for calculating the diffusion coefficient. It is a numerical solution [51–53]. By fitting the experiment, Huerke proposed an empirical formula for calculating the diffusion coefficient of CO_2 [54]. Unver and Himmelblau [55] studied the influence of temperature, pressure, concentration, brine salinity, and clay mineral content on the diffusion coefficient through experiments. Xu [56] conducted the experiments and found that the diffusion coefficient of CO_2 in natural gas was affected by the components of injected gas. He [57] simulated the diffusion law of CO_2 in different media through molecular dynamics. Du [58] studied the diffusion law of CO_2 in the crude oil-brine multiphase fluid and established the calculation model in the multiphase fluid. Kivi [59] studied the diffusion law of supercritical CO_2 in porous media considering the hydromechanical coupling effect. Liu [60] considered the diffusion of CO_2 flowing steadily in heterogeneous porous media, based on the mass conservation equation of CO_2-water two-phase flow, established a partial differential equation for CO_2 diffusion with velocity field, and verified the conclusion that CO_2 flows diffusion-steadily in heterogeneous porous media. Wang [61] studied the sealing characteristics, fluid migration, and transport characteristics, and related physical phenomena of CO_2 reservoirs with low porosity and permeability. The results show that the funnel-like diffusion halo can be formed by the combination of density difference, concentration difference and pressure difference in the process of supercritical CO_2 displacing saline solution. The physical model of CO_2 diffusion is shown in Figure 2.

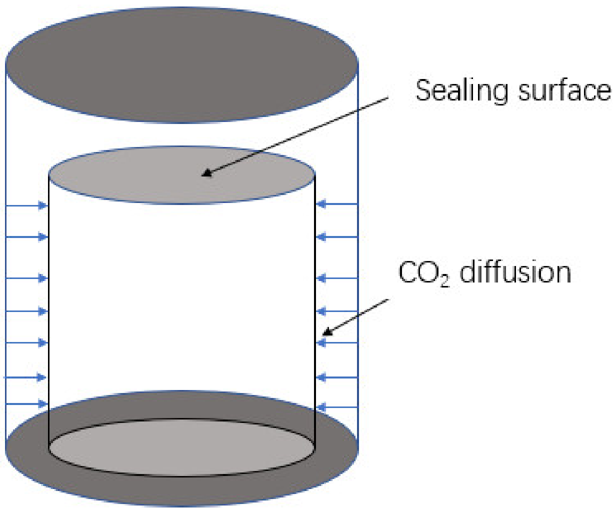

Figure 2. Schematic diagram of CO_2 diffusion physical model.

In terms of supercritical CO_2 migration law, Basbug [62] demonstrated through numerical simulation that CO_2 migration capacity and geological storage efficiency were greatly affected by reservoir heterogeneity, wettability, cap characteristics, and horizontal/vertical permeability ratio. Al-khdheeawi [63] studied the influence of brine salinity on the law of CO_2 migration based on the three-dimensional homogeneity model. In addition, Nordbotten and Dahle [64] established a supercritical CO_2 flowing model in the reservoir considering the action of capillary forces. Wang [65] applied the theory of mixture continuum to a mathematical model of thermal-fluid-solid coupling considering CO_2 migration in deep aquifers under the condition of CO_2 dissolution. Ao [66] conducted a supercritical CO_2 transportation experiment in shale and identified the adsorption-deformation characteristics of shale under the action of supercritical CO_2. Wu [67] conducted an experiment of supercritical CO_2 transport in coal seams, improved the D-R model, and described the law of adsorption and percolation of supercritical CO_2. Liu [68] conducted an experimental study on the flow migration of supercritical CO_2 and CH_4 binary system by CT visualization method. They analyzed the variation law of the apparent dispersion characteristics of supercritical CO_2.

In conclusion, there are many factors of the migration of CO_2 and gas mixture. In the process of CO_2 injection into carbonate sour gas reservoirs, the supercritical CO_2 will diffuse and migrate in the high-salinity brine and mixed gas. However, the laws concerning the diffusion, migration, and storage are not clear. The high temperature and high pressure lead the strong corrosion, extreme toxicity of H_2S and sulfur deposition, etc. Researchers around the world have not yet conducted experiments on the diffusion and migration of supercritical CO_2 in reservoirs, and complex fluids during the process of CO_2 injection and storage in carbonate gas reservoirs. Thus, the study on its mathematical model is rarely reported.

2.4. Recent Research on Mathematical Model of CO_2 Embedded Coupled Percolation in Carbonate Gas Reservoirs

Many researchers conducted mathematical models for percolation law in carbonate gas reservoirs. Kuo [69] established a one-dimensional radial flow model to study the effects of well spacing, gas production rate, and wellbore radius on sulfur deposition. Roberts [70] substituted solid sulfur deposition with oil components in the conventional black oil model. They set the relative permeability as zero to simulate the sulfur deposition process. Based on the literature of Gruesbeck and Cllins, Jamal and Abou-Kassem [71] established a mathematical model of elemental sulfur transport and deposition considering the deposition and adsorption of sulfur. Hands [72] established an analytical model for predicting sulfur deposition in fractured sour gas reservoirs. They considered the influence of near-wellbore temperature change and hydrodynamic effect in the formation. Civan [73] synthesized the damage caused by the migration, aggregation, deposition and blockage of particles in the pore throat of porous media to the reservoir and built the relevant mathematical model. Based on the conventional black oil model, Hu [74] analyzed the relationship between sulfur deposition and irreducible water saturation. They established the damage model of sulfur deposition to reservoir under non-Darcy flow. Mahmoud [75] studied the influence of sulfur adsorption on porosity, permeability, relative permeability, and rock wettability. Guo [15] proposed a new prediction model to calculate sulfur saturation in the near wellbore zone and analyzed the influences of gas reservoir temperature and pressure, gas viscosity, deviation coefficient, initial porosity, and absolute permeability on sulfur saturation. They also analyzed the influences of sulfur deposition on gas well productivity. The Chrastil model was further improved by variable constant method. A new model for predicting the solubility of elemental sulfur is proposed. Guo [76] established a three-dimensional multi-component mathematical model for sour gas reservoirs, and analyzed the effects of initial hydrogen sulfide concentration, rock permeability and airflow velocity on sulfur deposition. In addition, a reservoir permeability damage model is established that considered the effects of sulfur deposition, gas property changes, fracture development degree, and gas well production. The study found that fractured gas reservoirs with high H_2S content were mainly deposited in the near-wellbore zone, and fracture aperture had a significant impact on near-formation permeability. Guo [77] also established a temperature-pressure coupling model under the consideration of gas-sulfur fixation and gas-liquid sulfur integrated percolation and formed a reservoir-wellbore integrated simulation model.

Due to the high risk of fluid experiments in sour gas reservoirs, there are few studies on the microscopic percolation of sour gas. Wang [78] studied the microscopic percolation mechanism of gas and water in low porosity fractured sandstone gas reservoirs, taking fractured sandstone gas reservoirs in Kesen gas field, Kuqa Depression, Tarim Basin as an example. Based on rock samples and microscopic visualization techniques, saturated water-gas drive NMR and saturated gas-water drive CT scanning experiments were conducted. The seepage mechanism of the water invasion during natural gas development is simulated and the distribution of water saturation and the factors affecting driving efficiency are analyzed. Zhang [79] developed a testing device and method for microscopic percolation mechanism of sour gas reservoirs, studied the morphology and distribution characteristics of sulfur deposition in porous media during microscopic percolation of gas liquid sulfur, and explored the microscopic percolation mechanism of deep high temperature and sour gas reservoirs. Zhang [80] established the microscopic percolation mechanism experiment of sour gas reservoir, and studied the morphology, size, and distribution characteristics of sulfur deposition in porous media during the microscopic percolation of gas-solid sulfur and gas-liquid sulfur.

In terms of CO_2 storage, Bachu [81] proposed a CO_2 storage estimation model based on recoverable reserves, reservoir properties and in situ characteristics of CO_2 in depleted reservoirs. Zhao [82] established a CO_2 reservoir-storage potential evaluation model based on the shunting theory, thermodynamic theory and statistical analysis method. Li [83] studied the effect of CO_2 displacement and storage in sandstone reservoirs through numerical simulation and proposed a calculation method of CO_2 storage based on reservoir fluid physical properties. Jiang [84] established a reservoir CO_2 miscible flooding model based on the feature line method. According to the experimental results, Cheng [85] established a mathematical percolation model of CO_2 flooding in ultra-tight reservoir. Jiang [86] established a multi-component coupling percolation model of the continuous medium for CO_2 injection into shale with stratification method. Xu [87] used the triple pore reservoir model to evaluate the CO_2 storage in shale. Based on the theoretical CO_2 storage of gas reservoirs, Tang [88] considered the dissolution of CO_2 in brine and the interaction of CO_2–water–rock, established a calculation method for effective CO_2 storage of depleted gas reservoirs and established a mechanism model for simulation. Gao [89] proposed a CO_2 driven state storage calculation method based on component flash operation, which was used to calculate dissolved CO_2, bound CO_2, free CO_2, and total CO_2 storage potential based on the actual oilfield conditions and taking CO_2 drive storage mechanism into account. Zhao and Xin [90] measured the solubility of CO_2 under different temperature and pressure conditions and established a new formula for calculating CO_2 storage in combination with the numerical simulation method, aiming at the storage mode, temperature, and pressure of CO_2 in reservoir pore fluid. Dai [91] studied the effects of reservoir porosity, horizontal permeability, temperature, formation stress, ratio of vertical to horizontal permeability, capillary pressure, residual gas saturation, and other factors on reservoir CO_2 storage by applying the theories of seepage mechanics, multiphase fluid mechanics, and computational fluid mechanics. Cao [92] systematically analyzed the effects of CO_2 storage on depleted gas reservoir recovery, reservoir temperature, residual water saturation, and injection rate. In summary, although researchers have made some progress in coupled percolation model and CO_2 storage and utilization model for carbonate gas reservoirs, the coupled mathematical model of supercritical CO_2-brine-sour gas-rock percolation considering gas diffusion, flow migration and water–rock reaction has not been reported.

2.5. Rencent Research on CO_2 Injection to Enhance Oil Recovery Mechanism

CO_2 injection into low permeability reservoir has been an important means to improve oil recovery. CO_2 injection has the advantages of maintaining formation pressure, effectively reducing oil saturation pressure, oil volatility and oil viscosity, and expanding oil volume. Zhao [93] conducted laboratory experiments and numerical simulation studies on CO_2 flooding in ultra-tight reservoirs, and quantitatively analyzed the flooding mechanism of CO_2 in oil displacement efficiency. Combined with field test data, Liang [94] simulated and studied the mechanism of CO_2 flooding to improve oil recovery in tight reservoir. They optimized the development scheme of CO_2 flooding in tight reservoir. Guo [95] studied the optimal design of miscible and immiscible CO_2 flooding test scheme of Xinli Unit in Jilin Oilfield by high-pressure PVT cell and whole reservoir equation of State (EOS) simulation technology. The results show that CO_2-water injection after water flooding is an effective method to improve the recovery efficiency of a tight reservoir, which can significantly reduce the water production and improve the recovery efficiency of a tight reservoir. Vidiuk [96–98] established a component model for the injection of H_2S-CO_2 to study the mechanism related to the improvement of oil recovery by CO_2 injection, in which the increase in H_2S component can reduce the precipitation of asphaltene components in the reservoir and improve the fluid flow capacity in the reservoir. Heller [99–102] conducted supercritical CO_2 long core flooding experiments in gas reservoirs and the CO_2 injection desorption experiments in shale. The different properties between supercritical CO_2 and natural gas are beneficial to natural gas flooding and thus to improve natural gas recovery. The CO_2 injection can effectively improve the desorption rate and efficiency of adsorbed methane in shale. Based on the experimental results, Jikich [103–105] applied the numerical simulation model to study the mechanism of the CO_2 injection to improve the recovery of gas reservoirs. The study indicated that the storage of supercritical CO_2 as "cushion gas" can inhibit the breakthrough of bottom water and drive natural gas migration to upper parts of the gas reservoir. It also can help maintaining formation energy and improving the recovery of gas reservoirs.

The properties of supercritical CO_2 are very different from those multi-components natural gas. As a result, the supercritical CO_2 injection tends to accumulate at the bottom of the gas reservoir to form a "cushion layer", which can prevent the edge/bottom water from invading the gas reservoir. This layer also can maintain formation pressure and drive natural gas to the production well. Carbonate gas reservoirs in Sichuan Basin are characterized by deep burial, strong heterogeneity, high H_2S-CO_2 content, active edge/bottom water at a high temperature and high pressure. The mechanism of enhancing gas recovery by injecting CO_2 to form a "cushion layer". Whether it can prevent edge/bottom water intrusion is unclear. Recent theoretical and experimental studies cannot reveal the mechanism of CO_2 injection and enhanced gas recovery in carbonate gas reservoirs.

2.6. Potential Assessment of CO_2 Leakage

CO_2 leakage after storage has a significant risk to the environment. CO_2 leakage can be divided into sudden leakage and gradual leakage. Sudden leakage refers to CO_2 leakage caused by the fracture of injected depleted oil and gas Wells, resulting in sudden and rapid release of CO_2. Trapping CO_2 and other gases directly in geologic layers. Gradual leakage is the leaking of oil and gas through an undetected fault, fracture, or leak of the well, and its release slowly spreads to the surface. The leakage of CO_2 will have a significant impact on the atmosphere, groundwater, soil, etc., which may induce earthquakes [106].

The safety of CO_2 buried in strata is affected by many factors. The sealing property of structural trap and the long-term integrity of reservoir are two important conditions affecting CO_2 buried in strata. Pulchan [107] found that with the increase in CO_2 injection, reservoir pressure would increase, and fracture permeability would increase accordingly, resulting in the degradation of reservoir sealing property and CO_2 leakage. Li and Wu [108] designed a low temperature triaxial experimental device to study the strength of sediments with different hydrate saturations and established a new strength judgment basis. Duguid [109] collected data from existing Wells and used it to estimate the likelihood of a single well leaking and to estimate the economic loss caused by the leaking. In order to prevent the loss of CO_2 leakage, Li [110] designed a set of experimental system to explore the role of CO_2 hydrate in the secondary storage of leaked CO_2, studied the influence of CO_2 leakage on seawater PH value, and concluded that the formation of appropriate high-saturation CO_2 hydrate in the sediment above the leak point is conducive to the secondary storage of CO_2.

In summary, there are many factors affecting the leakage of buried CO_2. After CO_2 injection, the reaction between carbonate sour gas reservoirs and formation fluids leads to the decrease in geological sequestration strength, which may lead to the leakage of CO_2. Therefore, it is necessary to conduct research on the physicochemical properties of reservoir traps, and the optimal CO_2 injection rate according to different geological conditions.

3. Suggestions on Future Research Directions

In view of the main scientific and technical problems existing in the process of CO_2 injection storage and enhanced gas recovery in carbonate gas reservoirs, it is suggested to adopt the research methods of combining experiment and mathematical simulation, in both macro and micro scale. It is also suggested to conduct the following research.

3.1. Supercritical CO_2-Brine-Sour Gas Multi-Component Coexistence of Complex Fluid Phase Characteristics

During the process of CO_2 injection and storage, there is a phase change from the liquid phase in well to the supercritical phase in formation. In addition, the complex fluid with multiple components of supercritical CO_2, brine and sour gas in the formation has effects on dissolution and mixing between the fluids, which leads to the complex and variable phase behavior of the fluid. Understanding the phase change characteristics of supercritical CO_2-sour gas-brine multiphase complex fluid and obtaining the significant physical parameters of the fluid can guide the process and scheme design of CO_2 storage, which is of great significance for the scheme design and dynamic prediction of CO_2 storage. It is suggested to conduct phase characteristic experiment and thermodynamic model of phase equilibrium in supercritical CO_2-brine-sour gas multi-component coexistence systems.

3.2. The Intrinsic Mechanism of Supercritical CO_2-Brine-Rock Reaction

After CO_2 is injected into carbonate gas reservoir, the physical and chemical interaction between supercritical CO_2 and brine and rock will lead to the dissolution of some rock minerals and the formation of new precipitates. It will result in changes in the pore structure and mineral composition of the reservoir rock, also the physical and mechanical properties of the reservoir. In addition, considering liquid sulfur adsorption and sulfur fixation deposition, the existence of sulfur membrane on rock pore surface has a significant effect on the supercritical CO_2–brine–rock reaction process. According to the characteristics of mineral composition and physical properties of rocks in carbonate gas reservoir, it is necessary to conduct supercritical CO_2–brine–rock reaction experiment and study the interaction mechanism.

3.3. Diffusion Flow and Displacement Characteristics of Supercritical CO_2-Sour Gas

It is important to investigate the mixing behavior of CO_2 and natural gas in the process of injecting CO_2 for storage and enhancing gas recovery. At the same time, the adsorption and deposition of sulfur in pores will change the reservoir properties, and also have a certain influence on the process of supercritical CO_2 permeability. The diffusion and flow behavior of supercritical CO_2-sour gas was clearly defined. The CO_2 diffusion coefficient and the relative permeability curve of mixed gas were obtained. On this basis, the characteristics of mixed gas in the process of supercritical CO_2-sour gas displacement were studied, which was very important for the simulation study of CO_2 storage process and guidance of CO_2 storage and utilization of carbonate sour gas reservoirs. Recently, there are no relevant studies on the flow mixing behavior and phase permeability curve of supercritical CO_2-sour gas buried in CO_2 in carbonate gas reservoirs. Thus, it is necessary to conduct the study on the diffusion flow and displacement characteristics of supercritical CO_2-sour gas.

3.4. Machnism of Supercritical CO_2 Cushion Gas Prevents the Intrusion of Edge/Bottom Water

When supercritical CO_2 is injected into carbonate gas reservoir, due to the difference in fluid physical properties, CO_2 tends to accumulate at the bottom of the gas reservoir to form a cushion layer to prevent edge/bottom water from invading the gas reservoir. Due to the existence of sulfur deposition in the reservoir and the certain differences between the properties of sour gas and conventional natural gas, the expansion law of supercritical CO_2 in the reservoir is different from that of conventional gas reservoir. The understanding of spatial distribution characteristics is not clear. The mechanism of preventing edge/bottom water intrusion is not clear. Therefore, it is necessary to conducted research on the mechanism of preventing edge/bottom water intrusion by supercritical CO_2 cushion. It is suggested to conduct experimental studies on the solubility, diffusion coefficient and migration law of supercritical CO_2 under the condition of brine and gas, then develop a flat plate model to simulate the mechanism of CO_2 cushion to prevent edge/bottom water intrusion.

3.5. Evaluation of Injecting CO_2 Storage in Carbonate Sour Gas Reservoirs and Mechanism of Enhanced Recovery

By injecting CO_2 into the carbonate sour gas reservoir, not only can CO_2 storage be realized, but supercritical CO_2 injection can also improve the gas recovery. Considering that carbonate sour gas reservoirs are characterized by edge/bottom water, high temperature and high pressure, and sulfur deposition, identifying the mechanism of various factors on increasing the storage under different CO_2 sequestration mechanisms after supercritical CO_2 injection, inhibiting edge/bottom water breakthrough, maintaining formation pressure, and increasing reservoir percolation capacity can effectively guide the rational utilization of CO_2 injection and gas reservoir development. It is suggested to establish a coupled percolation mathematical model for carbonate sour gas reservoirs considering supercritical CO_2 diffusion, migration, and water–rock reaction. It will help study the mechanism of CO_2 injection storage and enhanced oil recovery in carbonate sour gas reservoirs.

4. Conclusions

In this paper, we conduct a literature review on the solubility and phase equilibrium thermodynamics of multi-component system of supercritical CO_2, and the recent research of supercritical CO_2-water–rock reaction experiment and coupling kinetic model, and the relevant research of diffusion migration and flow law of supercritical CO_2. The research status of coupled CO_2 percolation mathematical model of carbonate gas reservoirs and the mechanism of enhanced gas recovery also had been reviewed.

In view of the main scientific and technical problems existing in the process of CO_2 injection storage and enhanced gas recovery in carbonate gas reservoirs. It is suggested to carry out the following research in the future:

- Study on phase characteristics of complex fluid with multi-component coexistence of supercritical CO_2, brine, and sour gas.
- Study on the internal mechanism of supercritical CO_2–brine–rock reaction.
- Diffusion flow and displacement characteristics of supercritical CO_2-sour gas.
- Mechanism of supercritical CO_2 cushion to prevent edge/bottom water intrusion.
- CO_2 injection storage evaluation and enhanced oil recovery mechanism of carbonate sour gas reservoirs.

The mechanism of CO_2 injection storage and enhanced oil recovery of carbonate sour gas reservoirs at the later stage of development or depletion and abandonment is still unclear, and relevant basic research and field application technologies are still in the exploratory stage. Existing theoretical and experimental studies cannot reveal the mechanism of CO_2 injection for storage and enhanced gas recovery in carbonate sour gas reservoirs. It is necessary to combine physical simulation and mathematical simulation to carry out research.

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