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Numerical Study of Sensitivity Analysis on CO_2 Mineralization Sequestration in Basalt Reservoirs

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ABSTRACT

CO_2 mineralization sequestration in basalt reservoirs is an emerging and promising pathway for safe and effective CO_2 capture, utilization and storage (CCUS), contributing to carbon neutrality. However, the mechanism of CO_2 mineralization sequestration involves complex geochemical reactions, which are significantly affected by some factors. Mineral composition, temperature, pH, porosity, permeability and CO_2 injection rate are important factors that influence CO_2 mineralization in basalt, which require in-depth research and analyses. In this study, a radial model has been constructed using the multi-phase simulator GEM-CMG. This work focuses on the mechanisms of CO_2 mineralization and how these mechanisms affect the efficiency of CO_2 mineralization sequestration in basalt reservoirs. Sensitivity analyses are performed on the effects of injection mode (supercritical CO_2 injection and co-injection of CO_2 and water), injection rate and reservoir temperature on CO_2 transport and mineralization in basalt reservoirs. The results suggest that more than 60% of the injected CO_2 in basalt rocks mineralized within 450 days, and the majority of CO_2 was sequestered as magnesite and siderite. Compared to the supercritical CO_2 injection, co-injection of CO_2 and water can enhance dissolution trapping and thus accelerate the mineral trapping process. In addition, the injection rate has a major influence on the extent of CO_2-water-basalt reaction, and the CO_2 mineralization efficiency increases with the decrease of the injection rate. Within reasonable limits, higher reservoir temperature is more favorable to efficient CO_2 mineralization. This study is of great significance for the understanding of CO_2 mineralization mechanism in basalt reservoirs and provides valuable insights for optimizing CO_2 mineralization processes.

Keywords: CCUS, CO_2 mineralization, basalt, numerical simulation

NONMENCLATURE

Abbreviations

CCUS	CO_2 capture, utilization and storage
WMO	World Meteorological Organization
GCS	Geological carbon sequestration

Symbols

r	Reaction rate
$\hat{A}$	Mineral reactive surface area
k	Rate constant of the reaction
Q	Activity product of mineral reaction
K_{eq}	Mineral chemical equilibrium constant
k	Effective permeability
φ	Porosity

1. INTRODUCTION

The progress of industry and transportation enhances human's dependence on fossil fuels, which has caused a large amount of CO_2 emissions into the atmosphere, resulting in a series of climate issues such as global warming and rising sea levels. Based on data reported by the World Meteorological Organization (WMO), global temperatures in 2023 was around 1.45 ± 0.12 ℃ above 1850, significantly exceeding the temperature rise of the previous hottest year. It is further approaching the 1.5 ℃ temperature control target established by the Paris Agreement[1]. How to

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effectively reduce carbon emissions and achieve carbon dioxide capture and sequestration has become the focus of global scientists. Geological carbon sequestration (GCS) has become an important means of reducing CO_2 emissions and mitigating greenhouse effects on a global scale, which refers to the process of transforming CO_2 gas into a supercritical state through certain technologies, and injecting it into deep geological formations for long-term sequestration[2]. Under this sequestration system, the main sequestration mechanisms of CO_2 sequestration, arranged in descending order of sequestration safety, are structural trapping, residual trapping, dissolution trapping, and mineral trapping[3]. Among them, mineral trapping is the safest sequestration mechanism with the lowest risk of CO_2 leakage[4]. These mechanisms act sequentially with the increase of sequestration time. However, most rocks in conventional geological body (e.g., depleted oil and gas reservoirs, unexploitable coal seams and deep saline aquifers) have low reactivity and lack the divalent metal cations needed to form carbonate minerals, and thus mineral sequestration has limited areas for implementation and usually takes hundreds or even thousands of years[5].

Basalt is another promising geological body for CO_2 sequestration, which has a high percentage of ferromagnesian silicates and can rapidly convert CO_2 into stable carbonate minerals for long term CO_2 sequestration without the risk of CO_2 leakage, making it safer and more reliable than conventional CO_2 geological bodies[6]. In addition, basalt is widely distributed all over the world, with more than 5% of the land area and most of the oceanic crust composed of basalt, making it possible for large-scale utilization[7]. Currently, several engineering demonstration projects in the world are known to have conducted geological sequestration pilots of basalt CO_2, such as Carbfix in Iceland, Wallula in the United States, and Nagaoka in Japan, which have confirmed that the technology of CO_2 mineralization in basalt is feasible and has high development potential[8].

In recent years, many experimental and numerical studies on CO_2 sequestration in basalt have been conducted, and a deeper understanding of the mechanism, reaction rate and influencing factors has been gained. Matter et al.[9] conducted 1.4 m^3 CO_2 saturated water injection tests in a 230 m deep, 15℃ basalt reservoir in the Newark Basin, USA, demonstrating that basalt has a rapid CO_2 water-rock reaction rate under natural conditions. In addition, in CarbFix1, Iceland, Matter et al.[10] compared the difference between calculated and actually measured dissolved inorganic carbon content and ^{14}C isotopes. After more than 500 days of monitoring after the end of injection, they concluded that more than 95% of the injected CO_2 had been fully converted to carbonate minerals in less than 2 years, which is much more than expected. This finding suggests that CO_2 mineralization sequestration in basalt is much faster than in conventional sedimentary basin. Kanakiya et al.[11] experimentally investigated the microstructural changes in Auckland Volcanic Field basalts due to CO_2-water-basalt interactions after 140 days of reaction by measuring changes in parameters such as porosity, permeability, and pore geometry. Menefee et al.[12] designed a predictive modelling framework to assess the role of transport limitation and spatial distribution of minerals in fractured basalts exposed to CO_2 acidified fluids for mineral dissolution and carbonation reactions. Ratouis et al.[13] developed a one-dimensional reaction transport model using TOUGHREACT to study the injection of dissolved CO_2-fluid mixtures in a basalt geothermal reservoir. This work found that the mineralization process was influenced by mineral content, pH values between the injected fluid and the reservoir, mass fraction of injected CO_2 and the temperature of the injected fluid. However, fewer studies have been conducted on the long-term mineralization sequestration of large-scale CO_2 in basalt reservoirs, especially the relationship between parameters such as injection mode, injection rate and reservoir temperature with the mineralization reaction rate and sequestration efficiency.

The aim of this study is to establish a numerical simulation model to investigate the mechanism of CO_2 mineralization in basalt, and to analyze the effects of injection mode, injection rate and reservoir temperature on CO_2 mineralization efficiency. In this way, the results of the study are of great significance for the sequestration of CO_2 in basalt and provide a scientific basis for exploring efficient CO_2 sequestration solutions.

2. NUMERICAL MODELS AND METHODS

2.1 Numerical models

Over recent years, a large number of numerical simulation studies on CO_2-water-rock interactions and CO_2 mineralization in rocks have been carried out by many scholars. Numerical simulators applicable for studying the geomechanism of GCS include TOUGHREACT, NUFT, CHRUNCH, CHEMTOUGH, ECLIPSE, CMG-GEM as well as other simulators in which multiphase flow codes such as TOUGHREACT and CMG-GEM have been developed and used to study the carbon mineralization process related to GCS[14]. In this study, CMG-GEM was selected for simulation, which is a commercial software capable of simulating the subsurface transport, physicochemical processes of oil, gas and water under various complex geological conditions, and has been used for the study of CO_2 mineralization in basalt.

In this study, it is assumed that the flow in the gas and liquid phases obeys Darcy's law. The Peng-Robinson equation of state is used to simulate the fluid properties, the flow and transport equations are obtained based on the principle of conservation of mass and energy, and the parameters of CO_2 dissolved in the aqueous phase are calculated by Henry's law. In CMG-GEM, the reaction rate for mineral precipitation and dissolution is calculated by Bethke, 1996[15]:

$$r_j = \hat{A}_j k_j \left(1 - \frac{Q_j}{K_{eq,j}}\right), j = 1, \ldots, R_{mn}$$

Where r_j is the rate, $\hat{A}_j$ is the mineral reactive surface area, k_j is the rate constant of the reaction, Q_j is the activity product of mineral reaction j and $K_{eq,j}$ is the mineral chemical equilibrium constant, The ratio $Q_j / K_{eq,j}$ is the saturation index of the reaction. If $Q_j / K_{eq,j}$ >1, it means mineral precipitation occurs; if $Q_j / K_{eq,j}$ <1, mineral dissolution occurs. The reaction rate is zero when $Q_j / K_{eq,j}$ =1.

With mineral dissolution/precipitation, the effective permeability of the reservoir varies with porosity. In this study, the relationship between effective permeability and porosity is based on the Kozeny-Carman model[16]:

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

Where k is the effective permeability when the porosity is φ; k_0 is the effective permeability when the porosity is φ_0, k_0 and φ_0 are the original (initial) permeability and porosity.

2.2 Validation of the numerical simulation model

A three-dimensional model based on geological, hydrological and geochemical parameters from the site-scale Carbfix project in Iceland has been developed using the multi-phase simulator GEM-CMG. And the simulation results in Fig. 1 show that most of the injected CO_2 is converted to carbonate minerals around 1 year, which coincides with the time of carbon mineralization indicated by the monitoring results from the Iceland site. As can be seen from Fig. 2, the CO_2 mineralization is fast in the early stage of the simulation. Both siderite and magnesite are favorable carbonate precipitates. Siderite starts to dissolve about 0.5 year after the end of injection, and the released CO_3^{2-} and HCO_3^- are also gradually converted to magnesite by reacting with magnesium ions, which is in agreement with the phenomenon observed by Liu et al.[17] The CMG-simulated volume fraction of magnesite precipitation is about 90% for a period of 10 yr. while the result of liu et al. is about 88%. The two results are in good agreement, indicating that CMG can reasonably describe the geochemical reaction of basalt-CO_2-water.

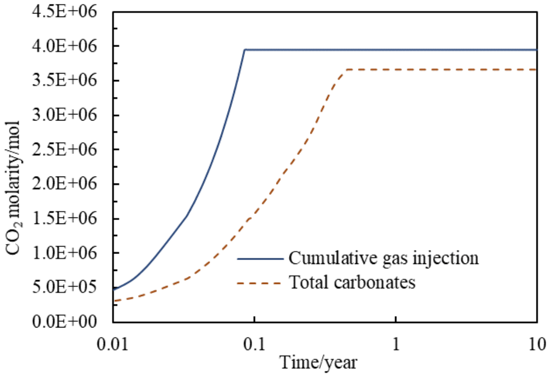

Fig. 1. Cumulative gas injection and total carbonate minerals over time

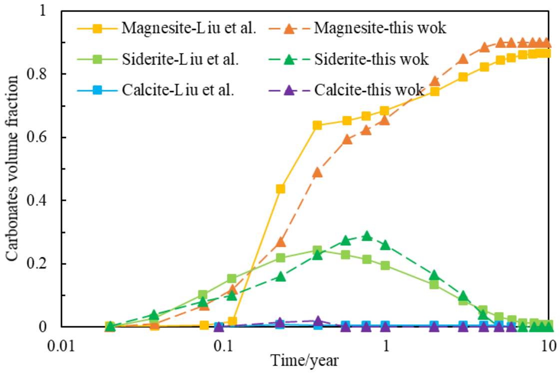

Fig. 2. Comparison of the results of this wok with that of Liu et al.[17]minerals over time

2.3 *Benchmark model*

Before determining the parameters of the benchmark model, several geological parameters of the major domestic and international basalt CO_2 sequestration sites were collected and summarized based on the contents of the literature research (Table 1)[18-24].

Table 1. Parameters of some pilot projects of basalt CO_2 sequestration

Basalt reservoir	Depth/m	Porosity	Permeability /mD	Temperature /℃	Injection rate/(m^3/day)
Iceland Carbfix1	400-800	0.1	300、1700	30-50	3100
Iceland Carbfix2	1900-2200	0.1	20-700	220-260	7000-15000
U.S. Wallula Project	828-887	0.15-0.25	75-150	35	20000
Leizhou Peninsula, China	-	0.03-0.15	-	-	-
Liaohe depression, China	-	0.03-0.22	0.01-10	-	-
Jiyang depression, China	2000-7000	0.06-0.1	0.06-1	-	-
Xujiaweizi depression, China	-	0.04-0.1	0.005-10	-	-

Considering that most of the basalt reservoirs in China belong to medium-high porosity-low permeability reservoirs, 0.1 and 20 mD are chosen as the porosity and permeability of the benchmark model.

Fig. 3 shows the schematic of the radial benchmark model. It consists of 100×30 grids in the radial and vertical directions, respectively. It is worth noting that the grid spacing increases isometrically in the radial direction to better observe the mineral dissolution/precipitation and CO_2 trapping after CO_2 injection into the basalt. The modelled reservoir has a thickness of 30 m, a depth of 800 m and an initial temperature of 40℃. The CO_2 injection point is located at the bottom center of the model. The gas injection rate is 5000 m^3/day for 2 months with a total simulation time of 10 years.

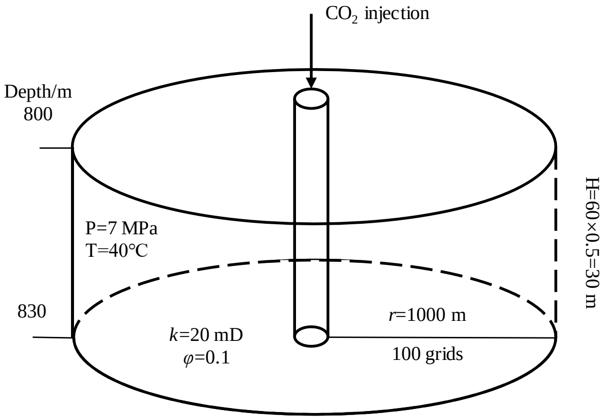

Fig. 3. Schematic diagram of the benchmark model

Table 2 provides more details about the parameters of this model. The initial hydrological data used in the simulations are presented in Table 3.

Table 2. Key parameters for the benchmark model

Parameter	Value	Unit
Depth	800	m
Temperature	40	℃
Pressure	7000	kPa
Irreducible water saturation	0.2	-
Compressibility	4.8×10^{-7}	1/kPa
Permeability	20	mD
Porosity	0.1	-
pH	9	-
Injection rate	5000	m^3/day
Injection time	2	month

Table 3. Initial aqueous phase compositions of the benchmark model

Chemical components	Concentration/ppm
Na^+	43
Ca^{2+}	5
Mg^{2+}	0.1
Fe^{2+}	0.1
Al^{3+}	0.054
SiO_2	24

Several common primary and secondary minerals in basalt reservoirs were selected, and their chemical reaction kinetic parameters were determined with reference to the existing literature (Table 4)[12,17,25,26]. Their respective initial volume fractions were set within a reasonable range. To better describe the precipitation of secondary minerals, the initial volume fraction of all potential precipitates in the simulation was set to zero. The mineral chemical reactions in basalt is shown in Table 5.

Table 4. Kinetic parameters for CO_2 mineralization[12,17,25,26]

Reaction location type	Mineral	Acid mechanism		Neutral mechanism		Basic mechanism		Initial volume fraction
		Log K_{25} (mol/m^2/s)	Ea (J/mol)	Log K_{25} (mol/m^2/s)	Ea (J/mol)	Log K_{25} (mol/m^2/s)	Ea (J/mol)	
Primary mineral	Albite	-8.86	67780	-12	66525	-15.6	297064	0.15
	Anorthite	-8.86	67780	-12	66525	-	-	0.19
	Fayalite	-6.85	92408	-12.8	94391	-	-	0.05
	Forsterite	-6.85	66944	-10.64	78993	-	-	0.05
	Diopside	-6.36	96106	-11.11	40584	-	-	0.1
	Hedenbergite	-6.36	96106	-11.11	40584	-	-	0.1
Secondarymineral	Magnesite	-6.38	14400	-9.34	23514	-	-	-
	Calcite	-0.3	14400	-5.81	23514	-	-	-
	Siderite	-3.74	36100	-8.90	62760	-	-	-

Table 5. Mineral chemical reactions in basalt

Mineral	Reaction
Albite	$NaAlSi_3O_8 + 4H^+ = Al^{3+} + Na^+ + 2H_2O + 3SiO_2$
Anorthite	$CaAl_2(SiO_4)_2 + 8H^+ = Ca^{2+} + 2Al^{3+} + 2SiO_2 + 4H_2O$
Fayalite	$Fe_2SiO_4 + 4H^+ = SiO_2 + 2Fe^{2+} + 2H_2O$
Forsterite	$Mg_2SiO_4 + 4H^+ = SiO_2 + 2H_2O + 2Mg^{2+}$
Diopside	$CaMgSi_2O_6 + 4H^+ = Ca^{2+} + Mg^{2+} + 2H_2O + 2SiO_2$
Hedenbergite	$CaFeSi_2O_6 + 4H^+ = Ca^{2+} + Fe^{2+} + 2H_2O + 2SiO_2$
Magnesite	$MgCO_3 + H^+ = HCO_3^- + Mg^{2+}$
Calcite	$CaCO_3 + H^+ = HCO_3^- + Ca^{2+}$
Siderite	$FeCO_3 + H^+ = HCO_3^- + Fe^{2+}$

3. RESULTS AND DISCUSSION

3.1 CO_2 mineralization in the benchmark model

Fig. 4 illustrates the cumulative CO_2 injection and the changes of trapping amount with time for various CO_2 trapping mechanisms. During CO_2 injection, structural trapping and residual trapping play a dominant role, with most of the sequestered CO_2 in a free state. After the end of injection, CO_2 is rapidly dissolved in formation water to form H_2CO_3. The proportion of dissolution trapping increases significantly, and H2CO3 further decomposes into H^+ and HCO3-. Then, the amount of mineralization sequestration increases with time because H^+ reacts with the main silicate minerals in the basalt formation, releasing divalent cations such as Mg^{2+} and Fe^{2+}, which in turn react with HCO^{3-} in the water to form carbonate precipitates. The speed of this mineralization reaction is relatively fast in basalt reservoirs, so most (more than 60%) of the injected CO_2 is converted into carbonate minerals within 450 days. Compared with conventional sedimentary basins where structural trapping and residual trapping dominate, the main mechanisms of CO_2 trapping in basalt are dissolution trapping and mineral trapping, which greatly reduce the time required to achieve the purpose of CO_2 sequestration.

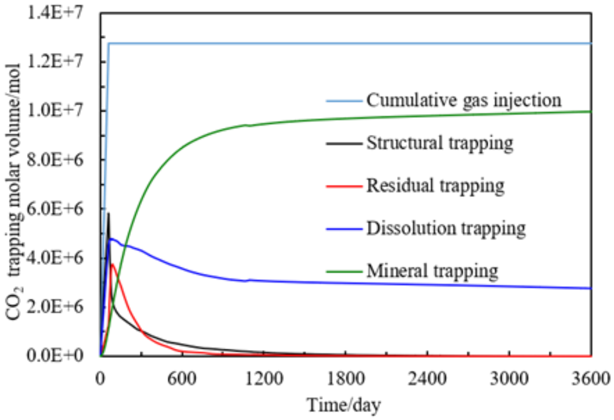

Fig. 4. Different types of CO_2 trapping over time

The variation of mineral molar changes in the model is shown in Fig. 5. Positive numbers are usually used to describe precipitation and negative values to describe dissolution. The main precipitating minerals are magnesite and siderite, and the dissolving minerals are forsterite and fayalite. This is due to the fact that forsterite and fayalite are actively reactive and susceptible to rapid dissolution in acidic environments, which provides Mg^{2+} and Fe^{2+} in favor of the precipitation of magnesite and siderite. As the simulation proceeds, more and more carbonates precipitate.

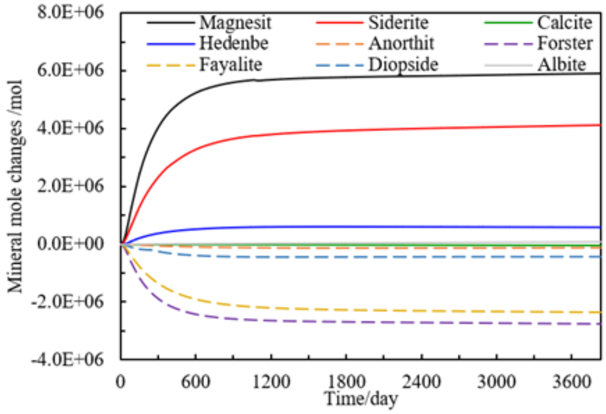

Fig. 5. The change of different minerals with time

3.2 *Effect of CO_2 injection mode*

Co-injection of CO_2 and water can enhance dissolution trapping and thus accelerate the mineral trapping process. In order to investigate the effect of injection mode on CO_2 sequestration, the simulation results of CO_2 and water co-injection were compared with those of supercritical CO_2 injection in the benchmark model. In the case of CO_2 and water co-injection, the CO_2 injection rate and injection time were the same in the benchmark model, the water injection rate was 200 m^3/day, and the ratio of CO_2 to water was 1:20.

For a comparative analysis of the extent of CO_2 mineralization, the following equations were used:

$$R_m = \frac{C_m}{C_i} \cdot 100\%$$

Where R_m is the CO_2 mineralization ratio; C_m is the amount of CO_2 mineral trapping, and C_i is the amount of cumulative injected CO_2.

As shown in Fig. 6, the CO_2 mineralization ratio is higher and the mineralization rate is faster in the CO_2-water co-injection scenario compared to the supercritical CO_2 injection scenario. In the CO_2-water co-injection scenario, the mineralization ratio reaches 60% in about 140 days, whereas in the supercritical CO_2 injection scenario it takes 450 days to reach this figure. This is because in the co-injection scenario, the CO_2 is almost completely dissolved in water at the moment of injection into the formation, thus directly carrying out the CO_2-water-basalt interaction motivated by dissolution trapping and mineral trapping (Fig. 7).

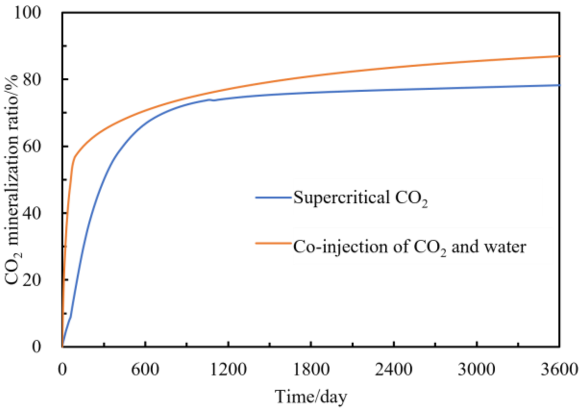

Fig. 6. CO_2 mineralization ratio of different CO_2 injection modes

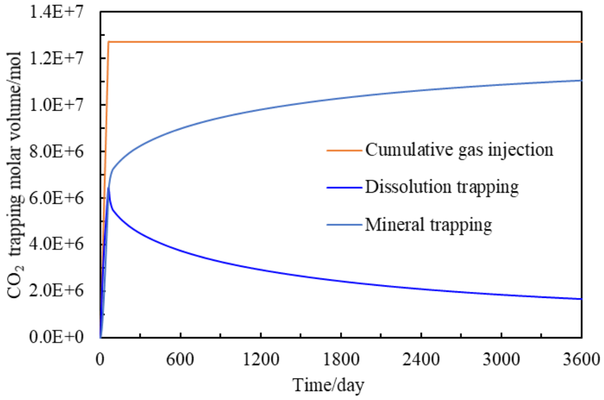

Fig. 7. Different types of CO_2 trapping over time

3.3 *Effect of CO_2 injection rate*

In addition to injection mode, the injection rate is also an important factor that affects the CO_2 mineralization ratio. In this section, the effect of injection rate on the CO_2 mineralization ratio was analyzed. The injection rates were set to 1000 m^3/day, 3000 m^3/day, 5000 m^3/day, and 7000 m^3/day, respectively. Fig. 8 shows the variation of the CO_2 mineralization ratio at different injection rates. It can be seen from the curves that the lower the CO_2 injection rate, the higher the mineralization efficiency, and this result is consistent with previous studies. The injection rate increases from 1000 m^3/day to 7000 m^3/day and the mineralization ratio decreases from 85% to 76%. The reason is that the proportion of CO_2 trapped by dissolution is higher and the CO_2 retention time in the pore space is longer at low injection rates, which amplifies the effect of the CO_2-water-basalt interaction. Fig. 9 demonstrates that the contribution of magnesite precipitation increases with the rise of injection rate, while the contribution of siderite is negatively correlated with the injection rate. As the injection rate increases from 1000 m^3/day to 7000 m^3/day, the contribution of magnesite to CO_2 mineral trapping grows from 55% to 68%, while the contribution of siderite decreases from 45% to 32%.

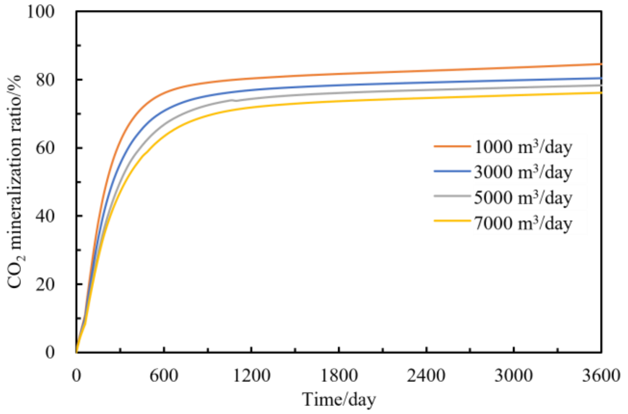

Fig. 8. CO_2 mineralization ratio at different injection rates

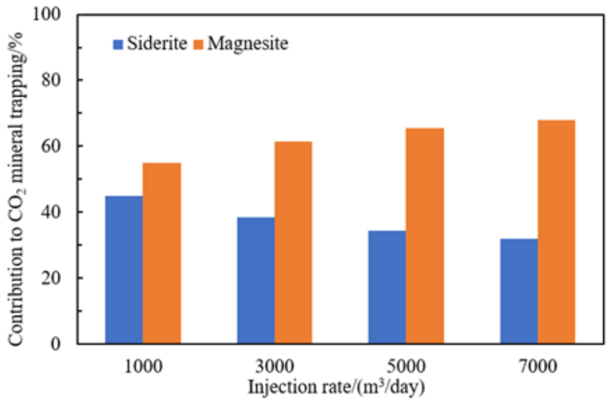

Fig. 9. Contribution of different minerals to CO_2 mineral

3.4 *Effect of reservoir temperature*

Different reaction rates of minerals lead to different extent of CO_2 mineralization. Experimental studies have shown that temperature is an important parameter affecting the reaction rate of CO_2 mineralization in basalt. Schaef and McGrail et al.[27] investigated the effect of temperature on the dissolution rate of basalt samples from the Columbia River, USA. They found that the dissolution rate increased with increasing temperature, and that decreasing the temperature from high to low decreased the dissolution rate of basalt by a factor of 100 over a temperature interval of 25-90℃. The results of Bénézeth et al.[28] also show that increasing the temperature helps the minerals in basalt to dissolve faster, which is conducive to increasing the amount of CO_2 mineralization. In this study, we set the temperature at 80 ℃, 60℃, 40℃ and 20℃ for 4 groups of models. Fig. 10 shows that the ratio of CO_2 mineralization increases with the rise of temperature. The CO_2 mineralization ratio at 80℃ is nearly doubled compared to that at 20℃. The reason is that the dissolution rate of ferromagnesian silicates is accelerated by the rise of temperature, which promotes the CO_2 mineralization.

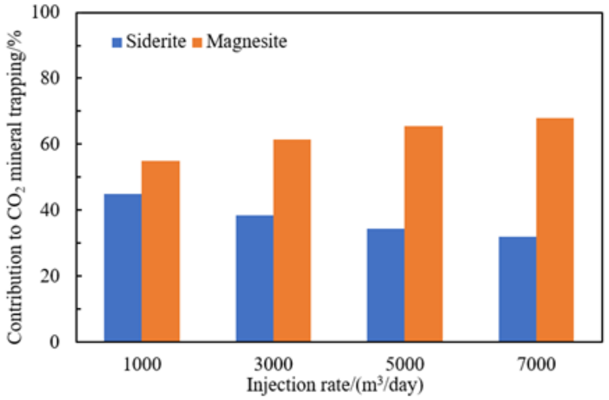

Fig. 10. CO_2 mineralization ratio of different temperature

4. CONCLUSION

One of the prospective methods used for safe and permanent sequestration of CO_2 underground is to inject it into basalt reservoirs. In this study, a numerical simulation method was used to investigate the mechanism of CO_2 mineral trapping in basalt reservoirs. On this basis, the effects of sensitive factors such as CO_2 injection mode, injection rate and reservoir temperature on CO_2 mineralization were analyzed. The following conclusions can be drawn from the analyses above：

(1) Basalt rocks are rich in a large number of highly reactive carbonate-fixing minerals, which can rapidly transform the injected CO_2 into stable carbonate minerals for long-lasting carbon fixation. The simulation results show that most (more than 60%) of the injected CO_2 is converted to carbonate minerals within 450 days after CO_2 injection. The dissolved minerals in basalt reservoirs are mainly forsterite and fayalite, while the precipitated minerals are mainly magnesite and siderite.

(2) Compared with the supercritical CO_2 injection, the CO_2 and water co-injection provides higher CO_2 mineralization ratio and faster mineralization rates. The reason is that the co-injection of CO_2 with water enhances the CO_2 dissolution trapping, and the resulting acidic environment is conducive to a rapid CO_2-water-basalt reaction. In addition, CO_2 injected by this method dissolves almost completely at the moment of injection into the formation, avoiding the potential problem of leakage.

(3) CO_2 injection rate is an important parameter affecting the extent of CO_2-water-basalt reaction. Furthermore, the simulation results illustrate that the

CO_2 mineralization efficiency increases with the decrease of CO_2 injection rate.

(4) Within a certain range, the increase in reservoir temperature helps dissolve some silicate minerals and accelerate the CO_2 mineralization. The results show that the amount of CO_2 mineralization at 80℃ is nearly doubled compared to that at 20℃.

DECLARATION OF INTEREST STATEMENT

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper. All authors read and approved the final manuscript.

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