

Using the precipitation process of potassium carbonate with Ethanol + KOH solution to enhance the efficiency of CO₂ mineralization storage in low permeability reservoirs

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ABSTRACT

Carbon dioxide displacement and storage is the most feasible technology to realize carbon neutralization, and also the key technology to improve the recovery of tight reservoirs. CO₂ capture and carbonization can be achieved by precipitation of potassium carbonate by using the "Ethanol + KOH" solution system. The reaction process in the solution system is affected by the ethanol concentration, resulting in different CO₂ carbonization amounts with the change of ethanol concentration. At the same time, the potassium-based acid salt generated by precipitation can react with water to complete the regeneration of ethanol. In this paper, experimental means are used to study the CO₂ capture efficiency of the "Ethanol + KOH" system, monitor the ethanol content in the solution in real time, and screen out the best ethanol concentration suitable for the formation temperature. Add KOH to the solution, use the ethanol regenerated in the solution after carbonization reaction to carbonize again, and determine the maximum CO₂ capture of "ethanol + KOH". Based on the high temperature and high pressure core displacement device, the CO₂ burial experiment after the injection of "Ethanol + KOH" solution was carried out to clarify the change rule of CO₂ burial under the action of "Ethanol + KOH" system in low permeability cores. The research results show that "96% ethanol+3g KOH" can effectively capture CO₂, and each capture will produce 4.56g precipitation on average. At the same time, after the core is saturated with "96% ethanol+3gKOH" solution, CO₂ is injected to generate sediment, and the core permeability decreases by about 15%. The research results of this paper prove that, compared with direct

injection of CO₂ into the formation, injection of this system into the formation in advance can accelerate the CO₂ carbonization process, thus effectively improving the CO₂ burial efficiency.

Keywords: Carbon dioxide, Ethanol+KOH+CO₂, potassium-based, CO₂ Storage

1. INTRODUCTION

Climate change is a global issue that has garnered significant international attention from governments and individuals worldwide^[1-3]. How to deal with excessive greenhouse gases and mitigate the greenhouse effect is a major challenge that human development needs to face^[4-6]. Carbon Dioxide Capture and Storage (CCS) technology is widely recognized as the most cost-effective and feasible approach for reducing CO₂ emissions and addressing the global warming challenges resulting from the combustion of fossil fuels^[7-9].

It can not only rely on a large amount of storage space in the formation to store greenhouse gases, but also use its own properties to change the properties of crude oil and improve its recovery rate^[10-12]. This process includes three steps: capture, transportation, and burial.

Carbon dioxide geological burial refers to storing carbon dioxide in adsorbed, water-soluble, and mineralized forms within buried geological structures^[13-16]. Currently, the main geological burial options include depleted oil and gas reservoirs, deep salt water reservoirs, unexplainable coal seams, and deep-sea burial^[17, 18]. According to different rock types and reservoir types, carbon dioxide injected into the ground can be stored underground through various burial mechanisms^[19-21]. The burial mechanism can be roughly divided into physical burial, geochemical burial, hydrodynamic burial and adsorption burial. Geochemical burial can be categorized as dissolution burial and mineralization burial.

Compared to China, foreign countries have significantly more mature CO₂ displacement and geological storage technologies^[22-24]. A large number of CO₂ displacement and geological burial pilot tests and field applications have been conducted in developed countries such as the United States and Canada^[25-27]. Currently, global CO₂ storage projects still maintain a rapid growth momentum. R. Kovscek have used numerical simulation methods to clarify the impact of CO₂ injection into water layers and oil layers on the geological storage of CO₂ and crude oil recovery rate. According to the study findings, the injection of CO₂ into both water and oil layers yields the most substantial crude oil recovery rate^[28]. Serdar Bender studied the impact of injected gas types on CO₂ geological storage and crude oil recovery. The research results indicate that since flue gas does not require purification and separation, injecting flue gas produces greater economic benefits than pure CO₂^[29]. Yao Zhenjie have established a CO₂ potential evaluation model for the Yanchang Oilfield in the Jingbian CO₂ experimental area. The research results indicate that the CO₂ storage capacity in the Jingbian CO₂ experimental area reaches $209.879 \times 10^4 \text{ t}$ ^[30]. Liu Bin evaluated the impact of reservoir heterogeneity on CO₂ flooding effectiveness. The research results indicate a positive correlation between crude oil recovery rate and reservoir heterogeneity. The stronger the reservoir heterogeneity, the worse the CO₂ flooding effect^[31]. Pufu Xiao conducted displacement experiments on four different injection schemes for Daqing ultra-low

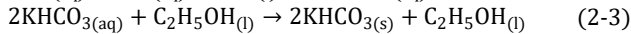
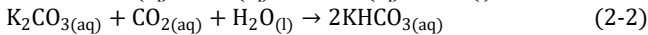
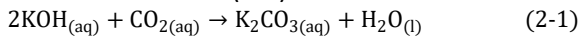
permeability oil reservoirs. The four schemes included CO₂/water alternating injection, CO₂ immiscible flooding, miscible flooding, and CO₂ huff and puff. The research results indicate that the remaining oil is mainly distributed in pores with pore sizes less than 10 ms, while CO₂ miscible and non miscible flooding mainly extract crude oil from large pores^[32]. Tang Yong conducted long core CO₂ displacement experiments to investigate the effects of high temperature and pressure on CO₂ enhanced oil recovery, specifically focusing on the changes in permeability and injection rate. The research results indicate that under the action of dissolution and gravity, the breakthrough of carbon dioxide lags behind, resulting in an increase in recovery rate^[33]. Jiang have established a multi-component numerical simulation considering multiple transport mechanisms (Kundsen diffusion, adsorption) to study the burial effect of CO₂ in shale gas reservoirs. The results show that the fracture properties have great influence on shale gas productivity. CO₂ injection into shale gas reservoir is a technically feasible method for CO₂ capture and enhanced recovery of shale gas. At the same time, N₂ can also be selected to assist CO₂ burial in shale gas reservoir^[34]. Zhao Renbao established a calculation model for CO₂ diffusion coefficient in bulk and porous media based on Fick's law. The research results indicate that as the viscosity of crude oil increases, the CO₂ diffusion coefficient decreases and the time for the system to reach equilibrium extends^[35]. Mu et al. used numerical simulation to study the effects of different impurities (N₂, SO₂) on the dissolution and burial mechanisms. The simulation results indicate that an increase in N₂ or O₂ concentration reduces the rate of CO₂ dissolution, weakens the ability to dissolve and store, and further leads to CO₂ leakage more easily through cracks and faults. On the contrary, SO₂ can advance the start time of density driven fingering, thereby promoting the dissolution and burial of CO₂^[36]. The research results of Zhang et al. show that during the CO₂ injection period, the burial of tectonics plays a major role, and the contribution of solution burial gradually increases with the amount of dissolved CO₂ in formation water. After CO₂ injection is stopped, tectonics traps and residual gas traps gradually decrease, solution traps increase, and mineralization burial contribution increases with time^[37].

The current research focuses on assessing the potential of CO₂ storage under different geological conditions, as well as the contribution of various burial mechanisms to CO₂ storage. However, it has neglected the slow process of CO₂ mineralization and storage,

which requires a long time. Therefore, this study independently built a CO₂ storage experimental device under formation temperature and pressure conditions, and conducted multi medium assisted CO₂ storage experimental research.

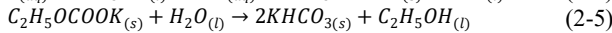
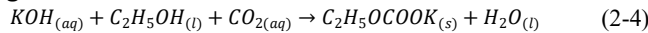
2. THEORY

Potassium hydroxide reacts with CO₂ to generate potassium carbonate (2-1). With the increase of CO₂ H₂O, potassium carbonate gradually generates potassium bicarbonate (2-2). Although potassium bicarbonate is soluble in water, it is insoluble in ethanol. The reaction is as follow (2-3).



When the concentration of ethanol in the solution is high, the (2-1) (2-2) (2-3) reaction is inhibited. KOH reacts with ethanol to generate potassium ethoxide, and finally KOH reacts with ethanol to generate potassium ethoxide, which is then converted into PEC and water, as shown in Formula (2-4) and (2-5).

At the same time, some PEC react with water in solution to complete ethanol regeneration and KHCO₃ generation.



To sum up, the mixed system of ethanol and KOH can produce KHCO₃ and PEC precipitation. The CO₂ injected is fixed through mineralization reaction. The type of sediment depends on the ethanol concentration in the solution^[38, 39].

3. EXPERIMENT SECTION

3.1 Experiment of carbon dioxide mineralization in ethanol + potassium hydroxide solution

Eight kinds of CO₂ mineralized solutions were prepared by dissolving 3 g KOH in 500ml ethanol aqueous solution with different concentrations (70%, 80%, 90%, 92%, 94%, 96%, 98%). Put the solution into a CO₂ reactor that can control the temperature. The flow chart of CO₂ mineralization reaction experiment is shown in Figure 1.

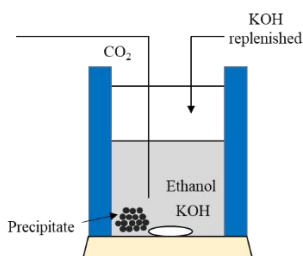


Fig. 1. Flow Chart of CO₂ Mineralization in Ethanol + Potassium Hydroxide Solution

(1) Inject CO₂ into the reactor and stir the solution at a speed of 380 rpm to mineralize CO₂.

(2) During the reaction process, use the pH meter in the reactor to measure the PH change every 1 minute.

(3) After the reaction, filter the carbonization solution to obtain the precipitate, and dry it at 50 degrees Celsius.

(4) Change the ethanol concentration and reaction temperature in mineralized solution, and repeat (1) - (4).

3.2 Experiment of carbon dioxide burial in cores after the action of ethanol+ potassium hydroxide solution.

The best proportion of CO₂ mineralization reaction is selected according to part 3.1 experiments. This article uses a self-developed CO₂ storage device to conduct CO₂ storage experiments and study the pore permeability changes of different permeability cores after saturation with a mixed solution.

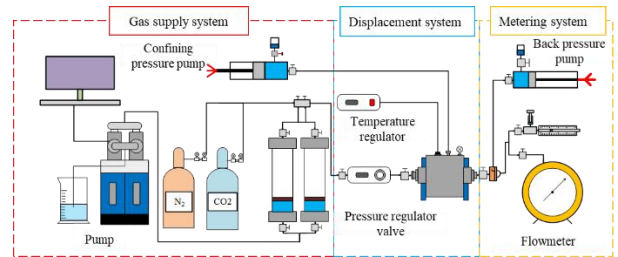


Fig. 2. Flow chart of core scale CO₂ storage experiment

(1) The core pore distribution under original conditions was determined using a nuclear magnetic resonance experimental device.

(2) Place the saturated CO₂ mineralized solution of the core in the core support to maintain the temperature of the core support at the formation temperature.

(3) The inlet end of the core holder injects CO₂ at a constant rate. Measuring core gas permeability and porosity.

(4) Saturated the core with water again, and use a nuclear magnetic resonance experimental device to determine the pore distribution after the CO₂ mineralization reaction.

4. RESULTS AND DISCUSSION

4.1 Experimental Research Results of CO₂ Capture

Experimental study on CO₂ capture was carried out under atmospheric pressure and 80 degrees Celsius. The CO₂ capture characteristics of dissolved 3g potassium hydroxide were determined at different

ethanol concentrations (75%, 80%, 85%, 90%, 92%, 94%, 96%, 98%). In accordance with the reaction process described in section 3.1, CO₂ was injected into the KOH + ethanol solution system to carry out the experiment on mineralization capture. The characteristics of the precipitation quality changing with ethanol concentration are shown in Figure 3, and the pH change characteristics of the reaction solution are shown in Figure 4.

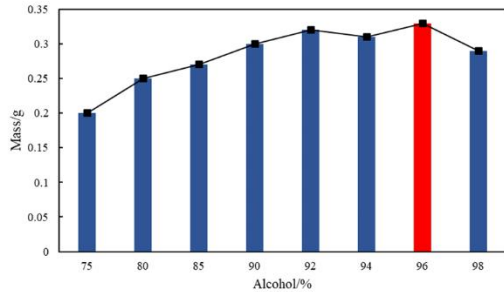


Fig. 3. Flow Chart of CO₂ Mineralization in Ethanol + Potassium Hydroxide Solution

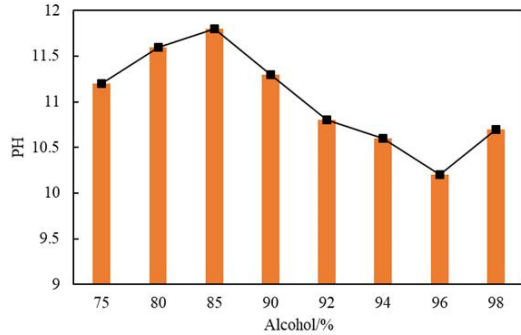


Fig. 4. Variation characteristics of PH value

The results show that the ethanol potassium hydroxide system can effectively carry out CO₂ mineralization and capture, thereby accelerating the CO₂ mineralization and capture process. Comparing the CO₂ precipitation results with different ethanol concentrations, it can be seen that the "96% ethanol+3 gram potassium hydroxide " 500 milliliter solution has the highest quality of CO₂ capture, so it is the best ratio of CO₂ mineralization capture solution.

4.2 Experimental Research Results of CO₂ Mineralization and Storage

The purity of CO₂ and N₂ used in the experiment is 99% and 99% respectively. The experimental liquid was compounded by 95% KOH and ethanol. The selected core porosity and permeability are shown in Table 1.

Table 4-1 Physical Property Data of Core Foundation

Core	Porosity /%	Permeability /mD	Porosity change rate/%	Permeability change rate/%
#1	13.2	6.18	7.57	12.13
#2	12.6	6.05	3.96	15.20
#3	11.2	5.83	3.57	10.63

#4	15.1	7.38	3.31	11.38
#5	13.6	6.32	3.67	10.60
#6	13.2	7.01	3.03	11.41
#7	12.2	6.35	4.91	12.12
#8	13.2	7.13	2.27	12.90
#9	12.8	6.11	6.25	12.43
#10	11.8	5.99	4.23	13.02
#11	10.82	5.01	4.73	13.7

Table 4-1 Physical Property Data of Core Foundation (continued)

Core	Porosity /%	Permeability /mD	Porosity change rate/%	Permeability change rate/%
#12	10.82	5.01	5.23	13.98
#13	10.76	4.95	5.73	14.04
#14	10.67	4.86	6.23	14.15
#15	10.16	4.35	6.73	14.21
#16	9.39	3.58	7.23	14.29
#17	8.83	3.02	7.73	14.46
#18	8.76	2.95	8.23	14.63
#19	7.83	2.02	8.73	14.81
#20	6.81	1	9.23	15.05
#21	6.31	0.5	9.73	16.12
#22	6.24	0.43	10.23	18.21
#23	6.04	0.23	10.73	20.16
#24	5.96	0.15	11.23	24.26
#25	5.91	0.1	11.73	26.45
#26	5.86	0.05	12.23	30.02
#27	5.84	0.03	12.37	34.26

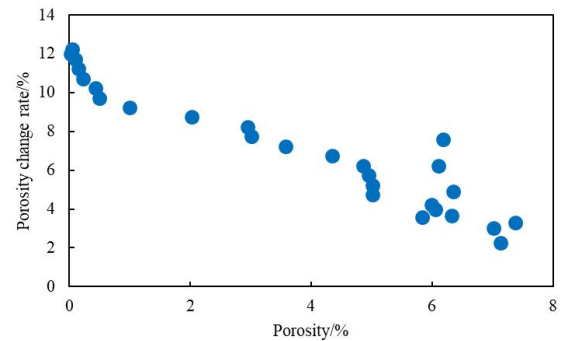


Fig. 5 Porosity change after CO₂ storage

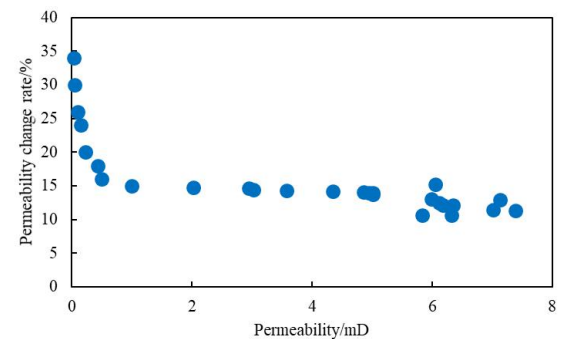


Fig. 6 Permeability change after CO₂ storage

Based on the experimental steps in section 3.2, CO₂ mineralization and burial experiments were conducted using sandstone cores. The results of CO₂ mineralization and burial experiments on low-permeability cores are shown in Figures 5 and 6. After CO₂ mineralization and burial, the core porosity decreases by an average of about 7%, and the permeability decreases by an

average of about 16%. The porosity and permeability change rate has a negative correlation with the porosity and permeability data. When the permeability is less than 1 millidarcy, the porosity and permeability change greatly.

4.3 Experiment Results of Nuclear Magnetic Resonance

Porosity and permeability are not sufficient to characterize the efficiency and conductivity of inter-reservoir pores in tight formations. Therefore, Nuclear Magnetic Resonance (NMR) experiments are frequently used to assess fluid mobility in various types of pores^[40,41]. The foundation of nuclear magnetic resonance is the interaction between the magnetism of atomic nuclei and the external magnetic field. The return of nuclei from a polarized state to the equilibrium state is referred to as the relaxation process, with the duration required being referred to as the relaxation time. The relaxation time can be divided into two types: longitudinal relaxation time T_1 and transverse relaxation time T_2 . T_2 is commonly used in the laboratory to characterize the depletion process of the sample. During the relaxation process, pore size plays an important role, and the relaxation rate is related to the rate at which protons collide with the surface. In large pores, collisions occur less frequently and the relative relaxation time is longer. It can be concluded that the size of pores is positively correlated with the relaxation time of hydrogen nuclei. For nuclear magnetic resonance experiments, the lateral relaxation time of rock samples measured in a uniform magnetic field can be expressed as:

$$\frac{1}{T_2} = \frac{1}{T_{2B}} + \frac{1}{T_{2D}} + \rho_2 \frac{S}{V}$$

T_2 is the lateral relaxation time of the rock, ms; T_{2B} is the lateral relaxation time of the fluid volume, ms; T_{2D} is the diffusion lateral relaxation time of the fluid, ms; ρ is the T_2 surface relaxation strength; S/V is the specific surface area of pores, $1/\mu\text{m}$.

This experiment clarified the characteristics of fluid changes in dense rock cores saturated with different solutions through nuclear magnetic resonance experiments. The research results are shown in Figures 7 and 8.

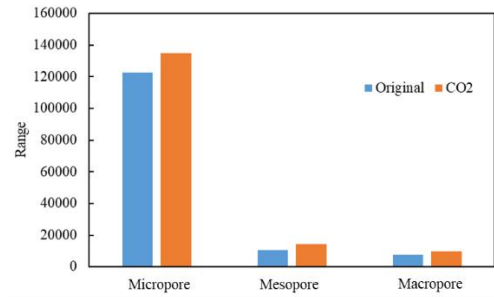


Fig. 7 Variation characteristics of different pores

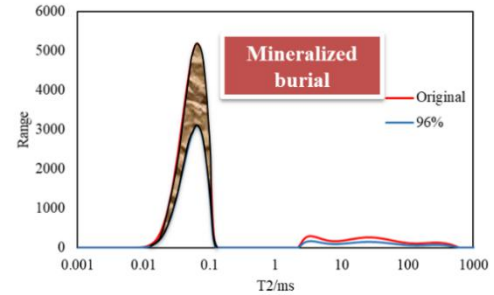


Fig. 8 Carbon dioxide burial NMR experiment(96% ethanol)

The experimental results show that after CO_2 treatment, the average pore size of the reservoir increases by about 12.8%, with medium and large pores increasing by 30% and micropores increasing by 12%. Injecting CO_2 into the reservoir will enhance pore connectivity and reduce the degree of CO_2 storage. From the analysis of Figures 8, it can be seen that conducting CO_2 burial experiments after saturating the core with an "ethanol+ potassium hydroxide " solution can effectively bury CO_2 in the formation in the form of sediment.

Due to the large number of small pores in the core, the contact area between CO_2 and solution is relatively large, there are more precipitates in the micropores (decreases by about 20%). It will be seen from this that the reservoir treated with " 96% Ethanol+3 gram " solution can significantly accelerate the efficiency of CO_2 mineralization and burial, shorten the time of CO_2 mineralization and burial, and increase the total amount of CO_2 burial.

5. CONCLUSIONS

(1) The research results show that the ethanol potassium hydroxide system can effectively carry out CO_2 mineralization and capture, thereby accelerating the CO_2 mineralization and capture process. It can be seen that the "96% Ethanol+3 gram potassium hydroxide " 500ml solution has the highest quality of CO_2 capture, so it is the best ratio of CO_2 mineralization capture solution.

(2) After CO_2 mineralization and burial, the core porosity decreases by an average of about 7%, and the permeability decreases by an average of about 16%.

The porosity and permeability change rate has a negative correlation with the porosity and permeability data. When the permeability is less than 1mD, the porosity and permeability change greatly. 96% Ethanol+3 gram potassium hydroxide can accelerate the CO₂ precipitation process in the reservoir and shorten the mineralization and burial time of CO₂ in the reservoir.

(3) The results of nuclear magnetic resonance experiments indicate that after the action of " 96% Ethanol+3 gram " solution, CO₂ can be effectively captured in the form of precipitates in the reservoir, with the largest degree of capture in the medium to large pores. From this, it can be seen that the reservoir treated with " 96% Ethanol+3 gram " solution can significantly accelerate the efficiency of CO₂ mineralization and burial, shorten the time of CO₂ mineralization and burial, and increase the total amount of CO₂ burial.

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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