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Effects of Carbonated Water Injection on Lacustrine Carbonates of Mupe Member, an Analog of the Oil and Gas Brazilian Pre-Salt Carbonate Reservoir

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Abstract

The study describes the complex process of dissolution, precipitation and mineralogical alteration caused by saline carbonated water injection (CWI) and its effects on the petrophysical properties (porosity and permeability) of limestone samples from the Mupe Member, composed of lacustrine microbialites, from the Upper Jurassic part of the Purbeck Group lower portion. These limestones have been studied as they are partial analog of the Brazilian presalt Aptian carbonates, the most important oil reservoir in Brazil that have been under CO₂ injection for oil and gas production. These reservoirs deliver large amounts of CO₂ that are reinjected into the formation, which given the high reactivity of carbonate rocks in the presence of carbonic acid generated by the reaction between CO₂ and water, can cause damage to the rock's pore space.

Introduction

An examination of CO₂ sequestration in carbonate aquifers demonstrated that either a permeability improvement or reduction can be obtained, since changes in rock properties are case-dependent because it is related to the pore distribution, brine composition, and thermodynamic interactions. CO₂ solubility in water, which increases with rising pressure and decreases with increasing temperature and salinity, is inversely proportional to brine's pH and is directly related to rock dissolution and porosity and permeability improvement potential. This dissolved material can, however, negatively impact permeability and porosity when mineral precipitation occurs (Albuquerque et al., 2023; Rabe et al., 2024).

The laboratory campaign was conducted by using samples from the the Purbeck Limestone Group (Upper Jurassic-Lower Cretaceous) is part of the Wessex Basin, located in southern England and northern France. The basal part of the group is composed of the Mupe Member (Upper Tithonian), which includes an accumulation of in-situ build-ups of microbial mounds (with thickness from about 0.5 to 4m - or 1.6 to 13ft - and high preserved primary porosity) within less-porous bedded inter-mound packstones-grainstones.

Methods

The experimental workflow consisted of the petrophysical characterization of six Purbeck carbonate plugs, with diameter of 1.5 in. (about 3.8 cm) and length between 1.6 and 2.8 in. (about 4–7 cm). To achieve the proposed objectives, four laminated/massive samples with very low permeability (<5 md) and two vuggy/microbial samples with very high permeability (>1,700 md) underwent laboratory tests carried out before, during, and after CWI. Chemical composition was determined by ion chromatography. Effluent fluid samples were collected at the system outlet to be analyzed by ion chromatography: One aliquot during the initial desulfated seawater injection (DSW), one for each VP of carbonated seawater (CSW) injected, and two after the injection of 10 and 20 cm³ of deionized water, respectively, at the end of the experiment. Ion chromatography was performed using Metrohm Ion Chromatograph's 940 Professional IC Vario and the columns Metrosep A Supp 5 150/4.0 for anion separation and Metrosep C 6 150/4.0 for cation separation. For the sample preparation, each sample was diluted (1:100 for cations Ca, Mg, Na, and K and 1:200 for anions Cl and SO₄), filtered, and injected directly into the chromatograph in a simultaneous analysis method.

Results of the CWI tests

Fig. 1 shows the difference between the amount (in mg/L) of calcium (Ca) and magnesium (Mg) that outflowed from the sample and the amount that was injected. Positive values indicate dissolution of calcium minerals (especially calcite) and magnesium minerals (possibly dolomite), and negative values indicate precipitation/loss of these ions from an aqueous solution. All samples indicated a high dissolution of Ca minerals, especially 3Y, in which more than twice the pore volume (PV) of the other samples was injected (Table 1). The dissolution of material from samples in which 5 PVs were injected (6Y, 6Z, and 1X) was very similar, despite the difference between the PV of each sample (Sample 1X has 40% more PV in comparison with 6Y and 6Z). The dissolution of the low-permeability samples that had higher PVs injected (6X and 3Z) was also similar (considering that 6X was injected with 2 PVs less than the 3Z). Comparing samples with similar permeabilities (6Y with 6X and 6Z with 3Z), the pressure difference stabilization after 5 PVs injected seems to have favored dissolution in the later stages of injection, generating a higher calcium output compared with the injected VP: An increase of injected PV in 16–23% generated an increment of 46–49% in Ca output and a permeability increase of 5.4–7.2 times (Table 1). Four samples (6Y, 6Z, 1X, and especially 3Z) showed precipitation of Mg minerals, and two samples (6X and 3Y) presented its dissolution (Fig. 1). These two may be due to the dolomitization that occurred in these samples (dolomite being a calcium and magnesium carbonate mineral abundant in nature). Fig. 2 indicates the difference between the amount (in mg/L) of chloride (Cl), sodium (Na), potassium (K), and sulfate (SO₄) that outflowed from the sample and the amount that was injected.

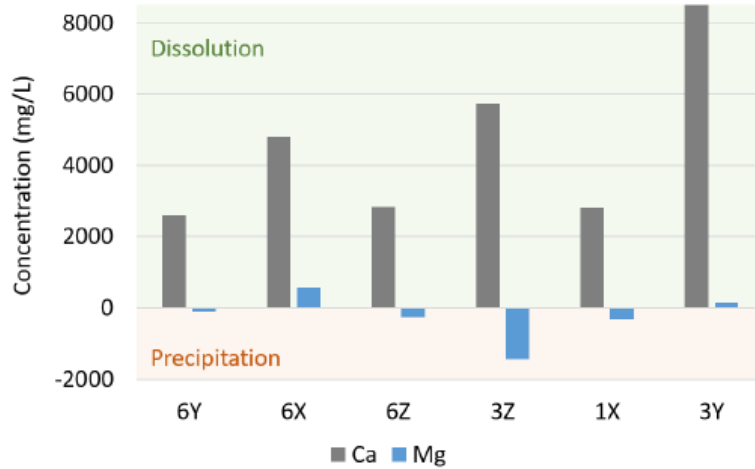


Figure 1. Difference between the amount (in mg/L) of Ca and Mg that outflowed from the sample and the amount that was injected.

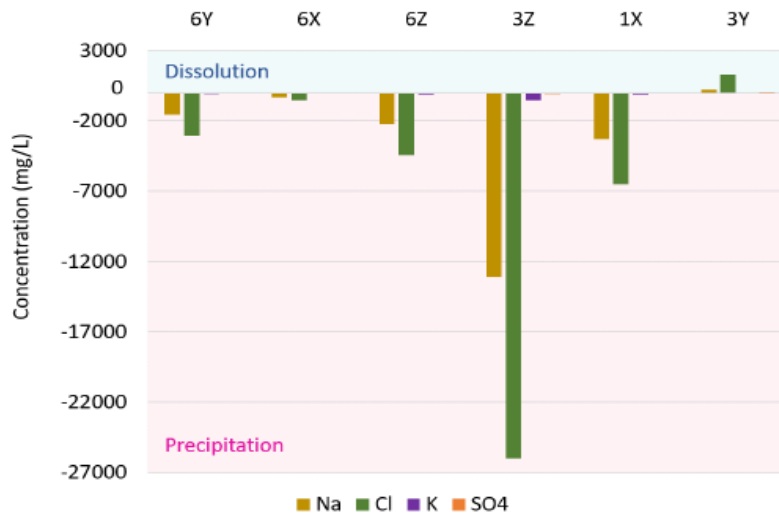


Figure 2. Difference between the amount (in mg/L) of Cl, Na, K, and SO4 that outflowed from the sample and the amount that was injected.

Sample		6Y	6X	6Z	3Z	1X	3Y
PV before CWI (cm ³)		5.63	4.18	5.60	3.63	7.89	8.37
PVs of CSW injected		5	8	5	10	5	10
Total volume of CSW injected (cm ³)		28.15	33.44	28	36.3	39.45	83.7
CSW injection duration (minutes)		28	61	66	47	62	111
Total calcium output (ppm)		2,591	4,818	2,845	5,749	2,826	8,518
Nitrogen permeability (md)*	Pre-CWI	0.63	0.19	1.53	3.31	1748.76	1795.41
	Post-CWI	6.99	10.51	5.44	64.27	1650.39	1603.78
	Δk	+1,006%	+5,433%	+256%	+1,845%	-5.6%	-10.7%
CSW permeability (md)	Pre-CWI	0.41	0.16	1.57	2.13	~2,000	227.26
	Post-CWI	6.90	9.61	4.13	32.82	Uncertain	303.01
	Δk	+1,583%	+5,906%	+163%	+1,441%	-	+33%
Helium porosity (%)	Pre-CWI	8.14	6.40	7.62	6.60	12.10	12.50
	Post-CWI	7.70	6.50	7.10	6.05	12.45	12.30
	$\Delta\Phi$	-5.4%	+1.6%	-6.9%	-8.3%	+2.9%	-1.6%

*Klinkenberg effect corrected (Klinkenberg 1941).

Table 1. Samples' PV before CWI, PVs and total volume of CSW injected, total calcium output, nitrogen and DSW permeabilities, and helium porosity before and after CWI

Discussion

Positive values indicate dissolution, and negative values indicate salt precipitation. Five samples indicated that salt precipitation may have occurred, mainly NaCl and, to a lesser extent, K and SO₄ salts, and 3Y showed little dissolution of these minerals. Salt precipitation was not directly related to the samples' porous volume, or the amount of CW injected, probably due to the pore structure of each one. Sample 3Z, being the most heterogeneous sample due to it having a contact between the vuggy/microbial facies and the laminated/massive facies, had the highest amount of precipitation. Since the samples were cleaned for salt removal after CWI (using the hot Soxhlet extraction method), it is assumed that salt precipitation did not change the post-injection analysis results.

Salt precipitation is an issue to be considered, especially in samples with high heterogeneity, mainly in contact between facies with different permeabilities. The CWI tests were executed with flow rate (1 cm³/min) likely impacted dissolution, particle migration, and salt precipitation during injection, being high for low-k samples and low for high-k samples. This is a factor to be considered in practical applications.

The considerable increase in permeability for the laminated/massive low-permeability facies samples together with the small change in the porosity values indicates good feasibility for the use of samples similar to those of the Mupe Member in CO₂ storage, especially in shallow reservoirs, with characteristics similar to those used during the experiments. For higher temperature and pressure settings, changes in CO₂ solubility must be considered, as should the potential for post dissolution carbonate redeposition once the injected cold fluid heats up to reservoir temperature.

For the vuggy/microbial facies samples, the small amount of formation damage generated can be a problem in large-scale applications. More studies on these rocks are needed to determine whether these results would preclude the application of CCS in similar formations.

Conclusions

The experimental results showed that samples with high permeability showed a small decrease in permeability, possibly indicating formation damage, while low-permeability samples presented a significant increase in permeability with little change in porosity, indicating feasibility for carbon capture and storage (CCS) in similar samples in analogous experimental conditions (20°C and 500 psi). For samples with more pore volumes injected, the pressure stabilization seems to have favored dissolution in the later injection stages, indicated by the highest output of calcium ions. In all samples salt precipitation was observed during injection, especially in the more heterogeneous rocks, presenting a possible formation damage issue.

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