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Laboratory Experiments on CO_2–Water–Dolomite Interactions for a Pilot CO_2 Storage Scheme in a Carbonate Reservoir in the Czech Republic †

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Abstract

This work investigates the geochemical response of carbonate reservoir rocks to CO_2-enriched fluids, focusing on mineralogical changes and potential geological storage consequences. Laboratory experiments involved Žarošice field (Czech Republic) dolomite rock and in situ representative pore water. The findings show that CO_2 injection causes carbonate and feldspar dissolution, causing a detectable porosity increase (~0.25 percentage points). These findings lay the groundwork for assessing the potential of using dolomitized carbonate rocks for pilot CO_2 sequestration projects in Central Europe.

Keywords: CCS (carbon capture and sequestration); reaction chamber; rock-solution-CO_2 interactions; carbonate reservoir; Vienna Basin

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1. Introduction

Lowering current atmospheric CO_2 concentrations is one of the major environmental challenges faced by the modern world. Carbon capture and storage (CCS) is a potential solution, consisting of CO_2 injection and safe long-term underground storage. CCS is highly dependent on reservoir rock properties such as porosity and permeability and mineral dissolution–precipitation processes [1].

Geological storage relies on several physicochemical mechanisms. Initially, structural, stratigraphic, or hydrodynamic trapping dominates in the beginning of project, while mineralization processes develop gradually after injection. These are influenced by fluid flow, capillary forces, convection, and molecular diffusion in both the liquid and gaseous phases. After the injection is complete, residual trapping, solubility trapping, and mineral reactions become critical. Dissolved CO_2 acidifies formation water, enhancing carbonate mineral dissolution and bicarbonate enrichment. The extent to which formations can bind CO_2 is a decisive technical and economic factor in CCS deployment [2–4].

This study is focused on carrying out laboratory experiments aimed at investigating the interactions between CO_2 and reservoir rock solutions under deep storage conditions to reproduce in situ pressure–temperature settings.

Although CO_2–carbonate interactions have been widely studied in international contexts, limited experimental data exist for Central European dolomitized reservoirs. The present study aims to bridge this gap by investigating the mineralogical and geochemical evolution of dolomite reservoir rocks under simulated deep storage conditions. The

objectives are to quantify mineralogical changes induced by CO_2-rich fluids, to assess dissolution and precipitation as a function of time, and to evaluate their implications for porosity evolution and storage integrity. We hypothesize that dolomite dissolution and secondary carbonate precipitation together control reservoir reactivity during prolonged CO_2 exposure.

The Žarošice oil field was discovered in 2001. According to geochemical research, the field is labeled as "mature", meaning it is in a phase of gradual decline and production is decreasing. With the decline in mining, options for utilizing the deposit for the geological storage of CO_2 are being explored. Žarošice is therefore one of the pilot sites for CCS in the Czech Republic.

2. Materials and Methods

This research involved collecting geological data, checking the quality of core samples, performing chemical analysis of solutions, and conducting laboratory simulations in a specially constructed reaction chamber.

2.1. Geological Characteristics

The Žarošice field is situated in southern Moravia, Czech Republic, in the transitional zone between the Western European Platform and the Carpathian thrust belt [5]. Its geological evolution is the product of a long and complex history dating back to the Mesozoic and Cenozoic eras of the Alpine orogenic cycle.

Regional lithospheric extension in the Early Jurassic initiated rifting and synrift basin formation, filled with coarse clastic sediments. It was followed by a tectonic regime of marine transgression, leading to the deposition of carbonate–siliciclastic sequences in passive continental margins. Uplifted rift shoulders and shallow-water environments developed platforms of carbonates, while the deeper basinal environments preserved organically rich marl units, such as the Mikulov marls [5–7].

In the Late Cretaceous, this area was a foreland of a synorogenic basin with deep-sea flysch sedimentation. This was followed by late and post-orogenic shallow marine and continental molasse deposition. Compressional Late Jurassic to early Miocene tectonics then resulted in thrusting of the Carpathian Flysch Belt over the European foreland. Concurrently, the Neogene Vienna Basin was developed as a foredeep overlapping with the thrust belt in part [7,8].

The Žarošice oil structure is a dolomitized buried reef dating back to the Jurassic period that has since been faulted and eroded partly from the west. Two different caprocks provide the seal, the Jurassic Marl of Mikulov Fm. in the eastern part of the field and Paleogene claystone in the western and northern parts, marked as Nesvačilka Fm. The reservoir rock mostly comprises heavily fractured dolomite of the Vranovice Fm. and, in the southern part near the base, dolomitic sandstones of the Nikolčice Formation.

Reservoir properties are a result of the combined action of depositional and diagenetic processes. The intercrystalline primary porosity of dolomites is usually 1–3%, and the porosity of a secondary origin, which is formed by fracturing and by dissolution, is 2–9%, being even higher in local zones. Permeability is 193–630 mD in accordance with the hydrodynamic tests [9].

Basal Jurassic sandstones and carbonate relict units around the northern edge of the Nesvačilka graben are considered the most favorable exploratory objectives for CO_2 storage. These are covered by thick low-permeability shales and mudstones that serve as effective caprocks and provide the required geologic conditions for secure long-term CO_2 storage [4,8,10].

2.2. Hydrogeological and Mineralogical Characteristics of the Site

The main rock material analyzed here consists of samples of dolomite from the Vranovice Formation (ZA4A borehole), retrieved from a depth of around 1500 m (Table 1). This formation dating back to the Upper Jurassic (~100 Ma) consists mainly of light gray dolomite.

Table 1. Basic characteristics for the samples of reservoir rocks.

Borehole	Vertical Depth (m)	Stratigraphy	Lithostratigraphy	Lithology
ZA4A	1528.5–1537.5	Upper Jurassic	Vranovice fm.	Light gray dolomite
ZA4A	1571.0–1578.8	Upper Jurassic	Vranovice fm.	Light gray dolomite

Formation water was obtained from the Žarošice 4A well (by partner MND a.s.) at a depth of ~1530 m and subsequently analyzed to characterize its hydrochemical composition (Table 2). The water is highly saline, with a total dissolved solids (TDS) content of approximately 24,600 mg/L, and displays a near-neutral pH of 7.1. The ionic composition is dominated by sodium and chloride, with significant contributions from bicarbonates, calcium, magnesium, potassium, and sulfate. Trace amounts of lithium, strontium, boron, and iron were also detected.

Table 2. Results of the chemical analysis of formation water from the Žarošice 4A well.

Parameter	Unit	Formation Water ZA4A
Ca^{2+}	mg/L	111.0
Mg^{2+}	mg/L	107.0
Na^{+}	mg/L	8570.0
K^{+}	mg/L	252.0
Mn^{2+}	mg/L	0.0
Fe_{total}	mg/L	0.4
NH_4^{+}	mg/L	32.6
Li^{+}	mg/L	3.2
Sr^{2+}	mg/L	19.2
B^{3+}	mg/L	43.0
Cl^{-}	mg/L	12,300.0
Br^{-}	mg/L	54.0
I^{-}	mg/L	43.0
HCO_3^{-}	mg/L	2580.0
SO_4^{2-}	mg/L	535.0
TDS	mg/L	24,590.0
pH	-	7.1

TDS, total dissolved substances.

The mineralogical composition of the studied reservoir rock sample is presented in Table 3. The porosity of the rock samples taken from the borehole ZA4A was 8%.

The hydrogeological and reservoir parameters of light gray dolomite from Vranovice Fm. were measured by MND a.s. (Table 4). Permeability data obtained via hydrodynamic testing show anisotropic properties with a horizontal permeability of 500 mD and vertical permeability of 1600 mD. This type of anisotropy is characteristic of fractured carbonates and has important implications for fluid migration and the development of CO_2 plumes upon injection. Reservoir pressures at depths of approximately 1500 m are in the order of ~116 bar and the temperatures are around 53 °C. Under these conditions, the injected CO_2 will be in a dense phase with a density of 195 kg/m^3 and a viscosity of 1.5×10^{-4} Pa.s.

These physical properties are important input data for models of CO_2 behavior during injection and storage.

Table 3. Mineralogical composition of the studied rock sample.

Mineral	Borehole	Unit wt%	Unit vol.%
Dolomite	ZA4A	55.04	50.60
Ankerite		17.95	16.56
Calcite		-	-
Quartz		4.01	3.68
Muscovite		2.88	2.76
Kaolinite		3.12	3.23
Microcline		7.66	7.36
Albite		7.92	7.39
Chlorite		1.42	1.12

wt%, percentage by weight. vol.%, volume percentage.

Table 4. Hydrogeological and reservoir parameters and values of CO_2 density and viscosity for the Vranovice formation (data provided by MND a.s.).

Parameters	Unit	Value
Horizontal permeability k_R	mD	500
Vertical permeability k_Z	mD	1600
Thickness	m	200
Pressure P	bar	116
Temperature T	°C	53
Porosity	%	8.00
Maxsalinity S	g/L	72
CO_2 density	kg/m^3	195
CO_2 viscosity	Pa.s	1.5×10^{-4}

2.3. Methods

The methodological framework of this study was designed to evaluate mineralogical and geochemical modifications in carbonate reservoir rocks exposed to CO_2 injection under simulated deep storage conditions. A multi-stage approach was used, integrating petrophysical, mineralogical, and hydrogeological analyses with controlled laboratory experiments.

XRD analysis and reaction chamber experiments were performed in laboratories belonging to the Department of Geological Engineering at VSB-Technical University of Ostrava. The chemical analysis of reaction solutions was conducted by the Department of Environmental Engineering at VSB-Technical University of Ostrava.

2.3.1. Mineralogical and Petrophysical Characterization

The mineralogical composition of core samples was established via X-ray diffraction (XRD) analysis with a Bruker AXS D8 θ–θ powder diffractometer (Bruker AXS GmbH Karlsruhe, Germany) with a Bragg–Brentano geometry, operating with CoKα radiation (λ = 1.7903 Å, 34 kV, 30 mA). Quantitative phase analysis was used to quantify changes in bulk mineralogy prior to and subsequent to experimental exposure to CO_2-rich fluids.

The porosity was determined with the help of other techniques. Baseline values were obtained from acoustic well logging data provided by the operator (MND a.s.), and high-resolution measurement was performed by means of mercury intrusion porosimetry (Autopore 9220, Micromeritics Instrument Corp., Norcross, GA, USA) from cylindrical

plugs cut from drill cores. This dual tactic allowed both the large-scale and pore-scale porosity evolution to be studied.

2.3.2. Reaction Chamber

Serial experiments were carried out in a specially manufactured high-pressure reaction chamber (VŠB Technical University of Ostrava, Faculty of Mining and Geology, Ostrava, Czech Republic) designed to replicate reservoir conditions. This device consisted of two independent stainless-steel cells (inner diameter: 141 mm; length: 368 mm) located in a pressure-resistant cylindrical vessel (outer diameter: 169 mm; length: 955 mm). The vessel was equipped with DN150 PN160 blind flanges, a protective internal lining inert to CO_2, and safety coatings to prevent corrosion. Heating was provided by an external coil, while a rocking mechanism enabled controlled oscillatory motion (two cycles per day), simulating dynamic geohydrodynamic conditions.

The experiments were conducted at 150 bar pressure and under temperatures of less than 53 °C, consistent with in situ conditions at a depth of around 1500 m. The reaction medium was Žarošice 4A well formation water. Samples of rock were retained for 30, 90, 120, and 180 days in separate cells to allow for temporal assessment of water–rock–CO_2 interactions.

The temperature and pressure in the reaction chamber were constantly monitored by using digital sensors. CO_2 was introduced through a precision mass-flow controller to ensure constant composition throughout the whole experiment. Periodical calibration fully confirmed that fluctuations in temperature and pressure did not exceed 1% during the 180-day test. A schematic representation of the experimental setup, including gas inlet, heating, and sampling lines, is provided in Figure 1.

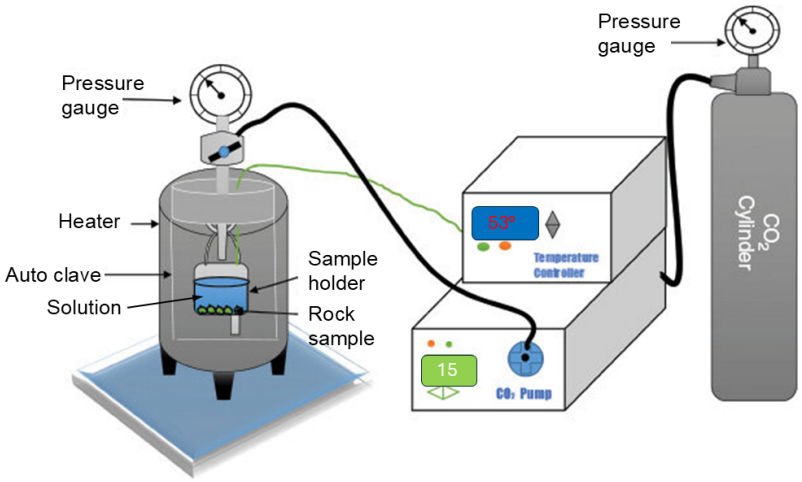


Figure 1. Schema of the reaction chamber.

2.3.3. Hydrochemical Analysis

After every experimental interval, the aqueous solutions were measured to determine the geochemical changes. Chemical analyses of the samples were carried out in the laboratory facilities of the Department of Environmental Engineering, VSB-Technical University of Ostrava. Ion chromatography and atomic absorption spectrometry determined major cations and anions, and trace elements were analyzed via ICP-OES. These data served to assess mineral dissolution/precipitation trends and to cross-check mineralogical findings from XRD.

3. Results and Discussion

The experimental program provided a detailed dataset describing the mineralogical evolution of dolomite reservoir rocks exposed to CO_2-rich fluids under simulated storage conditions. Changes were monitored through sequential XRD analyses of solid phases and the hydrochemical characterization of reaction solutions at 30, 90, 120, and 180 days. The results point to a dynamic interaction between mineral dissolution and precipitation, which together influence reservoir reactivity and storage behavior. The chemical analysis of formation water is presented in Table 5.

Table 5. Chemical analysis of reaction solutions after completing the experiments in the reaction chamber.

Parameter	Units	Input Analysis	ZA4A (30 Days)	ZA4A (90 Days)	ZA4A (120 Days)	ZA4A (180 Days)
pH	-	7.1	6.7	6.9	7.2	6.8
ρ at 15 °C	g/cm^3	1.0	1.0	1.0	1.0	1.0
Conductivity	mS/cm	36.5	36.9	35.3	37.2	38.0
K	mg/L	252.0	257.0	256.0	463.0	334.0
Na	mg/L	8570.0	4570.0	6260.0	8750.0	6770.0
Mg	mg/L	107.0	99.6	100.0	94.8	92.7
Ca	mg/L	111.0	285.0	315.0	348.0	702.0
NH_4	mg/L	32.6	9.1	15.8	19.4	19.1
Li	mg/L	3.2	2.8	2.9	2.8	2.7
Sr	mg/L	19.2	17.0	13.6	16.3	13.4
Mn	mg/L	0.0	0.9	1.1	0.7	0.4
Fe	mg/L	0.4	0.01	94.9	18.9	60.8
B	mg/L	-	45.8	43.0	40.8	41.6
SO_4	mg/L	535.0	640.0	660.0	550.0	540.0
Cl	mg/L	12,300.0	13,250.6	12,727.6	13,046.7	12,444.0
HCO_3	mg/L	2580.0	2702.3	3440.4	3225.7	3660.0

3.1. Evolution of the Reservoir Rock

Mineralogical changes in the rock samples from borehole ZA4A can be assessed using the following table (Table 6). These changes occurred during the experiments in the reaction chamber.

Table 6. Mineralogical changes in the rock samples from borehole ZA4A 30, 90, 120, and 180 days after CO_2 injection.

Time	0 Days	30 Days	90 Days	120 Days	180 Days
			(wt%)		
Dolomite	55.04	36.33	42.43	63.62	36.45
Dolomite Ca-Fe	17.95	27.12	21.32	11.84	26.39
Quartz	4.01	7.71	5.78	2.93	4.49
Calcite	0	7.42	8.66	1.87	6.49
Muscovite	2.88	2.9	3.38	4.01	1.79
Kaolinite	3.12	4.32	1.43	2.27	4.13
Microcline	7.66	6.04	7.15	8.19	9.75
Albite	7.92	8.21	10.03	5.33	9.74
Chlorite	1.42	0	0	0	1.69

wt%, percentage by weight.

3.1.1. Thirty-Day Interval

In the initial 30 days, significant dissolution of the dolomite occurred, along with a measurable rise in Ca^{2+} and HCO_3^- levels in the solution (Table 6). This is taken to indicate congruent dissolution of the dolomite, which contributed to the accumulation of bicarbonate. The reducing concentration of Mg^{2+}, and the simultaneous increase in iron concentration, is consistent with the precipitation of Fe–Mg carbonates, most likely ankerite. Muscovite and albite remained unchanged mineralogically, but chlorite was entirely dissolved, bringing the resulting solution into contact with Mn^{2+}. Calcite precipitation, which explains the rise in Ca^{2+} concentration, is the result of the re-equilibration of saturation of the carbonate system under acidic CO_2-induced conditions.

3.1.2. Ninety-Day Interval

After 90 days, further dissolution of dolomite, kaolinite, and microcline was observed. Meanwhile, chlorite was completely dissolved. These reactions occurred in parallel with high concentrations of Ca^{2+}, Mg^{2+}, iron, and manganese. Calcite precipitation was simultaneously maintained, indicating a buffering reaction to progressive acidification. Albite contents increased, in spite of a significant decrease in Na^+ concentrations, and may reflect recrystallization or the selective preservation of feldspar phases. Muscovite showed consistent stability. These findings suggest that secondary carbonate formation may partly compensate for primary mineral dissolution and hence moderate porosity development.

3.1.3. One-Hundred-And-Twenty-Day Interval

At 120 days, a notable increase in dolomite content was documented, interpreted as either reprecipitation or stabilization associated with ankerite dissolution. The dissolution of ankerite released Mg^{2+}, Ca^{2+}, Fe, and HCO_3^-, which again promoted local calcite precipitation. Elevated Na^+ concentrations indicated the dissolution of albite, kaolinite, and quartz. A sharp increase in K^+, coupled with the precipitation of muscovite and microcline, suggests that alternative K-bearing phases (e.g., sylvite or carnallite) may have acted as sources of mobile potassium, not captured by XRD analysis. Chlorite remained absent, indicating its consistent instability throughout the experiment.

3.1.4. One-Hundred-Eighty-Day Interval

By 180 days, further dissolution of dolomite and muscovite was observed, leading to the further release of Ca^{2+}, Mg^{2+}, and K^+ into the solution. The concurrent precipitation of calcite and dawsonite suggests a reorganization of carbonate mineral phases. Secondary dolomite and siderite will begin to precipitate within 6 months of CO_2 injection as the primary carbonate mineral, ankerite, dissolves from the rock matrix (Figure 2). Minor increases in quartz, kaolinite, albite, and microcline contents were observed, while Na^+ concentrations declined, reflecting albite precipitation.

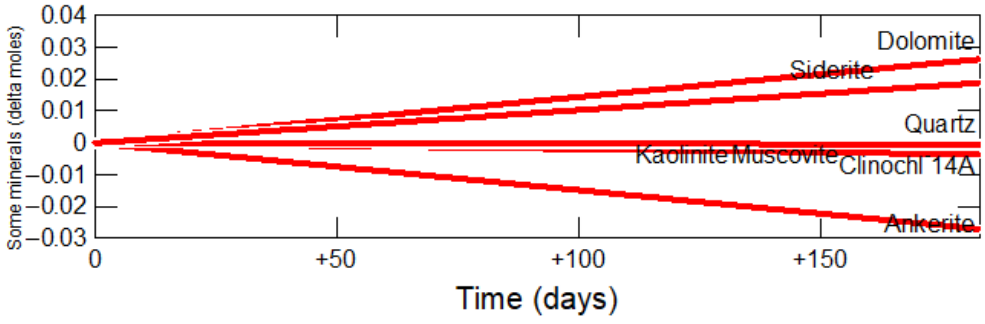


Figure 2. Changes in mineral composition during the CO_2 injection due to ankerite dissolution.

4. Conclusions

The laboratory experiments of CO_2–water–dolomite interactions presented here offer new information concerning the geochemical and mineralogical reactions of carbonate reservoirs under the relevant deep geological storage conditions. The main results can be outlined in the following way:

Dynamics of precipitation and dissolution of minerals—Dolomite was always susceptible to dissolution when it was exposed to CO_2-containing fluids, mostly in the beginning of the experiments. This was accompanied by the precipitation of calcite and ankerite, the most common secondary carbonates. The observed oscillations in dolomite and calcite contents between the beginning and after 180 days likely reflect alternating dissolution–precipitation dynamics controlled by transient pH and ion concentrations. During early exposure, rapid CO_2 dissolution increased fluid acidity, promoting dolomite breakdown and calcite saturation. As equilibrium was established (120 days), secondary dolomite reprecipitated from bicarbonate-enriched solutions.

Reactivity of the silicate and clay minerals—Chlorite was completely unstable across all time intervals, providing the dissolved phase with iron and manganese. Kaolinite and microcline showed limited dissolution, while muscovite and albite were generally stable, with solubility of muscovite occurring only over a longer time interval (180 days).

Development of the chemical composition of fluids—The gradual enrichment of the solution with Ca^{2+}, HCO_3^-, Mg^{2+}, and iron and the declining Na^+ concentrations are indicative of a close relationship between carbonate dissolution and secondary precipitation processes.

The present results align with previous studies addressing carbonate reactivity under CO_2-rich conditions. Luquot & Gouze (2009) [2] evidenced that porosity and permeability evolution in flow-through carbonate experiments are driven by dissolution–precipitation feedback, while Izeg et al. (2008) [3] presented similar geochemical fingerprints of dolomite dissolution and calcite reprecipitation upon CO_2 flooding. Our long-term batch experiments complement these findings by emphasizing the temporal dimension of mineral reactivity, showing oscillatory alternations in dolomite and calcite contents from beginning to 180 days. All of these studies altogether emphasize the dynamic nature of carbonate buffering processes and their implications for CO_2 storage stability.

In summary, this research supports the perception that the Czech Republic's dolomitized carbonate reservoirs are potential candidates for pilot-scale CO_2 storage. The mineralogical changes observed here underscore the need for continued field and laboratory monitoring and long-term experimental research to better model reservoir development over time. Ongoing research will broaden these experimental studies to other potential localities, including the Uhřice oil field, to develop a more general geochemical context for CO_2 storage in European carbonate environments.

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