

# Micro-scale Insights into the Effects of Ionic Strength on CO<sub>2</sub> Induced Carbonate Rocks Dissolution

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## Abstract

Deep saline aquifers are appropriate for long-term storage of captured CO<sub>2</sub>[1]. Low pH environments of deep aquifers due to CO<sub>2</sub> injection and dissolution would trigger rock dissolution while, the extent of rock dissolution depends on the brine chemistry[2]. Carbonates are the main mineral forming the matrix of carbonate rocks[3]. Moreover, carbonate minerals show higher kinetic reactivity in comparison with alumino-silicate minerals, and therefore, these are the first minerals that react with acidic fluids. Our previous study showed that the brine chemistry affects the extent of carbonate mineral dissolution and consequently, the micro-scale structure and porosity ( $\Phi$ ) of the reservoir rock[2].

Even though it is well known that the aqueous phase in geological reservoirs has ionic strengths ranging up to several hundred g/L, the role of this parameter on geochemical rock-fluid interactions is still a subject of debate in the literature[2], [4] due to the variety and complexity of the employed rock and fluid compositions. Thus, this research aims to ascertain the relationship between the alteration of micro-scale pore structure of Indiana limestone, as a common carbonate reservoir rock (>98% calcite) and change in ionic strength of reactive CO<sub>2</sub>-saturated brine. Three static experiments were designed to evaluate the effect of ionic strength on the amount of CO<sub>2</sub>-saturated brine induced rock dissolution, using various analytical methods. Brines were synthesised by the most common types of ions[5] which are found in deep aquifers including Na<sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, Ca<sup>2+</sup> and Cl<sup>-</sup>, to reach ionic strengths from 0 to 0.6 mol/L. Indiana core plugs (with dimensions of 5mm×12mm in diameter and length, respectively) were initially equilibrated with brines for 25 days, based on geochemical modelling. Then CO<sub>2</sub> was injected into hydrothermal batch vessels at a pressure of 130 bar and temperature of 60°C that are representative of subsurface reservoir conditions. Experiments with CO<sub>2</sub> saturated brines were continued for 14 days to ensure that desired rock-fluid interactions have occurred. Brine samples (2 mL) were collected at each step to analyse the

Ca<sup>2+</sup> concentration using inductively coupled plasma-optical emission spectroscopy (ICP-OES) technique.

Micro-computed tomography ( $\mu$ -CT) scanner was used as a non-invasive methodology for obtaining information on the spatially resolved three-dimensional pore structure of the rock. Images were acquired with a 4  $\mu$ m spatial resolution and X-ray beam of 80 keV and 80 $\mu$ A before and after each experiment to evaluate the changes induced by CO<sub>2</sub>-saturated brine. The alterations of macro-porosity, interparticle sub-resolution micro-porosity and solid phase of the rock were assessed by  $\mu$ -CT images. The results of the ICP-OES were used to compare the amount of released Ca<sup>2+</sup> in the brine with the calculated volume of dissolved rock by  $\mu$ -CT images. Overall, results of ICP-OES and  $\mu$ -CT images illustrate that the extent of rock dissolution and alteration of pore structure of rocks were depending on the ionic strength of the solutions. Therefore, higher ionic strength leads to more rock dissolution. Good agreement was also observed between the total porosity calculated from  $\mu$ -CT images ( $\Phi = 0.203$ ) and helium porosity measurements ( $\Phi = 0.215$ ).

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