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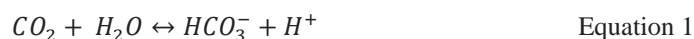
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### Abstract

One of the key technologies for reducing CO<sub>2</sub> from atmosphere is the Geological carbon storage (GCS) [1], and among the subsurface reservoirs deep saline aquifers are the ones that present the largest capacity for storing the CO<sub>2</sub> [2]. The dissolution of injected CO<sub>2</sub> in the formation brine will release proton (H<sup>+</sup>) and acidify the storage environment according to Equation 1. Therefore, the reactivity between rock and fluid will get more intense which emphasizes on the requirement of particular consideration to achieve a secure GCS project/operation.



Although it has been proved that different ion types are effective in determining the behavior of different phases at the rock-fluid interface [3], only a few studies have focused on the impact of ion type on mineral dissolution [4, 5] and thus there is a research gap regarding cation-induced rock dissolution. Furthermore, the NaCl solution is normally utilized for mimicking the formation brine and predicting the rock-fluid interactions at laboratory based experiments [6]. However, more CO<sub>2</sub> gets dissolved in presence of Na<sup>+</sup> in comparison with that dissolved in the presence of other monovalent cations, hence leading to over-estimating the reactivity during the experiment [7]. Therefore, the aim of this work is to evaluate the influence of the different ion types on rock-fluid interactions and the mechanisms behind the cations induced rock dissolution under high pressure-high temperature (HPHT) conditions that are similar to those found at aquifers. Since many of GCS projects are conducted at carbonate reservoirs [8], Indiana limestone was selected as the rock phase for this study. The synthesized solutions consist of the two most abundant monovalent

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cations exist in deep saline aquifer including  $\text{Na}^+$  and  $\text{K}^+$  to be in direct contact with Indiana limestone at static hydrothermal batch vessel for 14 days. To be representative of HPHT condition at GCS reservoirs a heat jacket was installed to set the temperature at  $60^\circ\text{C}$  ( $140^\circ\text{F}$ ) and injecting  $\text{CO}_2$  to build up the pressure up to 130 bar (1885 psi). Our primary hypothesis is that various cations' characteristics e.g., hydrated radii, bare radii and surface charge densities can describe variant level of accessibility for injected  $\text{CO}_2$  to the negatively charged rock surface due to the electrostatic repulsion/attraction forces and in consequence the initiation of rock dissolution. To investigate this hypothesis, various analytical techniques were carried out at different spatial scales (pore and core) to figure out the influence of various compositions of the solution on the mineralisation process of newly entered  $\text{CO}_2$ . The techniques include Powder X-ray Diffraction (PXRD), micro-Computed Tomography (Micro-CT) and Inductive Coupled Plasma-Optical Emission Spectroscopy (ICP-OES).

Based on the micro-CT images, dissolution occurred more uniformly at the outer surface of the sample volume; this is expected for static hydrothermal batch experiments. By comparing the results of ICP-OES after each experiment, the level of rock dissolution for each single cation can be manifested. Measurement of cation concentrations by ICP-OES before and after each experiment shows that NaCl brine is more reactive than KCl solution.

Moreover, the experimental conditions were simulated using PHREEQC to predict in-situ variable parameters due to the geochemical reaction phenomena during  $\text{CO}_2$  storage. The geochemistry modelling results depicted good agreement in terms of the dissolved species with the experimental data.

As the conclusion, this research work delivers new discernment on the functionality and impact of each ion type that presents in the fluid phase and mechanisms behind the reactivity among Indiana limestone and brine under GCS conditions. The results illustrate raising of rock-fluid interactions due to reduction of ion radius and each cation has its own role on determining the fate of  $\text{CO}_2$  at saline aquifers.

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*Keywords:* Deep Saline Aquifers, Geochemical Rock-Fluid Interactions,  $\text{CO}_2$  Injection into Carbonate Reservoirs

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