



**The Effects of Wet Supercritical CO₂ Treatment on the Storage Potential of Zululand
(South Africa) Sandstones.**

PhD Thesis

Prepared by

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Submitted to

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April 2021

Abstract

Understanding the mineral and microstructure changes in sandstone samples during wet supercritical CO₂ gas treatment is an important aspect of geological CO₂ sequestration. In order to gain insight into geological storage of CO₂ in South Africa, five core samples extracted from the reservoir caprock and separate sandstone aquifers within the Zululand Basin were studied. The samples were treated with CO₂ gas streams at typical reservoir temperature and pressures (120 bar and 45 °C, and 175 bar and 73 °C) under wet conditions. High pressure Parr reactors were used for storing the rock-gas-water mixtures for up to two months using a water:rock ratio of 23:1. Tests were conducted using pure CO₂ and a 99% CO₂ and 1% SO₂ mixed gas stream. Due to the prohibitive costs associated with CO₂ purification, the knowledge of the consequences of key impurities relevant to geological sequestration is critical.

Pre-and post- CO₂ /CO₂-SO₂ treatment characterisation was conducted using X-ray Diffraction (XRD) analyses, Fourier transform infrared spectroscopy (FTIR), and low-pressure gas adsorption (LPGA). In line with literature in the field, varying mineral alterations were observed in all CO₂ treated samples, mainly comprising of calcite, plagioclase and smectite dissolution and the precipitation of quartz, plagioclase, calcite and smectite. The formation of dissolution pores and pore clogging was indicated. In line with XRD results, increased microstructure heterogeneity was indicated in the FTIR profiles after CO₂ treatment through reduced peak intensities. Increases in the adsorption capacity, surface area and pore volume were observed in all samples except for the ZA sample, which showed contradicting results. This was attributed to the significant quartz precipitation observed in the sample.

Two samples (ZC and ZG) were further treated with CO₂-SO₂ gas mixture. After treatment with CO₂-SO₂ gas mixture, increases in mineral reactivity were observed in the ZC sample along with gypsum precipitation, indicating potential improvement in the lateral seal's self-sealing capacity. The introduction of SO₂ lead to the increase in quartz and plagioclase precipitation and increased dissolution of smectite and stilbite in the ZG sample. Minor changes in the pore structure were reported. The study presents a novel investigation of the changes expected to take place during CO₂ injection in sandstone basins. The documented findings provide a crucial foundation for the prediction of long-term CO₂ sequestration effects in the Zululand Basin of South Africa.