



# An experimental investigation of coupled chemico-mineralogical and mechanical changes in varying-cemented sandstones upon CO<sub>2</sub> injection in deep saline aquifer environments



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## ABSTRACT

Although CO<sub>2</sub> storage in deep saline aquifers is now accepted as a potential option for atmospheric CO<sub>2</sub> mitigation, the chemico-mineralogical property alterations in the aquifer formation associated with CO<sub>2</sub>/brine/rock mineral interactions, the corresponding influence on formation hydro-mechanical properties and the effect of rock mineral structure, are not yet fully understood. This study was therefore conducted to obtain a comprehensive understanding of the effect of long-term CO<sub>2</sub> exposure on the chemico-mineralogical structure and corresponding strength characteristics of saline aquifer rock formations using silicate cement (SS) and carbonate cement (CS) Hawkesbury sandstone samples collected from the Sydney basin. Sandstone samples were first reacted with brine+CO<sub>2</sub> under different injection pressures (both sub-critical (4, 6 MPa) and super-critical (8, 10 MPa)) under a constant temperature of 35 °C. A comprehensive chemico-mineralogical analysis (ICP-AES and XRD) was first conducted on both the rock mass pore fluid and the rock matrix over the saturation period of one year, giving special attention to the alteration of dominant rock minerals (quartz, calcite and kaolinite). The overall influence after 12 months of saturation with brine and CO<sub>2</sub> on the strength characteristics of the two types of sandstones (SS and CS) was then investigated and correlated with the chemico-mineralogical reaction, in order to understand the coupled process.

According to the test results, compared to the silicate cement-dominant mineral structure (SS), the presence of a carbonate cement-dominant mineral structure (CS) in the aquifer rock formation creates more significant alterations in the formation's chemico-mineralogical structure upon CO<sub>2</sub> injection. This is because calcite mineral reactions occur at much greater rates compared to quartz mineral reactions in CO<sub>2</sub>-exposed environments. In addition, although some minor precipitation of kaolinite minerals may also occur upon CO<sub>2</sub> injection, the effect may not be significant. Overall, rock mineral changes in deep saline aquifers upon CO<sub>2</sub> injection have a significant influence on the strength characteristics of the reservoir rock mass, depending on the aquifer mineral structure, and CS formations are subject to much greater strength property changes upon exposure to CO<sub>2</sub> than SS formations. Interestingly, CO<sub>2</sub> injection causes a strength gain in SS sandstone and a strength reduction in CS sandstone. These mechanical property alterations in aquifer rock formations are also dependent on the CO<sub>2</sub> injection pressure and phase, and increasing the injecting CO<sub>2</sub> pressure significantly enhances the changes, due to the highly acidic environment created by the enhanced CO<sub>2</sub> solubility process. Changing the CO<sub>2</sub> phase from sub- to super-critical condition also accelerates the reaction mechanisms, due to the greater chemical potential of super-critical CO<sub>2</sub>. However, overall SS sandstone exhibits more stable chemical-mineralogical and mechanical characteristics upon CO<sub>2</sub> injection than CS sandstone, and exhibits more suitable characteristics for CO<sub>2</sub> sequestration.

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## 1. Introduction

Over the past century, the emission of anthropogenic CO<sub>2</sub> into the Earth's atmosphere has significantly increased with the expansion of the industrial sector [18]. According to the International Energy Agency's (IEA) [13] report, even with concentrated action by the United Nations Framework Convention on Climate Change (UNFCCC) to limit temperature increases to under 2 °C, fossil fuels will still provide 60% of the world's primary energy demand by 2040 [13]. Due to the abundant use of fossil fuel combustion, such as coal, oil and natural gas, the levels of atmospheric carbon dioxide (CO<sub>2</sub>), an important greenhouse gas that modulates global climate change, have increased. To date, around a 35% increment (anthropogenic CO<sub>2</sub> emission is about 4% of the total yearly world CO<sub>2</sub> emission) in the CO<sub>2</sub> level has been recorded in the heavily industrialized areas of the world [19]. In addition, over the past century, the atmospheric CO<sub>2</sub> level has increased by more than 39% from 280 ppm during pre-industrial times to the record high level of 400 ppm in May 2013 [14]. Moreover, the Emission Database for Global Atmospheric Research revealed that the recorded global CO<sub>2</sub> emission value of 33.4 billion tonnes in 2011 was 48% higher than that of the value recorded two decades ago [12]. Table 1 summarises the amount of CO<sub>2</sub> emitted from main anthropogenic sources in 2011 [12,13].

In order to overcome this crucial issue of ever-increasing atmospheric CO<sub>2</sub> levels, many mitigation options have been considered [18], including 1) Improved energy efficiency and promotion of energy conservation, 2) Promotion of the use of low carbon fuels, including hydrogen and nuclear power, 3) Increased use of renewable energies, such as solar, wind, hydropower and bio-energy, 4) Introduction of geo-engineering approaches, such as afforestation and reforestation, 5) CO<sub>2</sub> capture and utilization (CCU), and 5) CO<sub>2</sub> sequestration or CO<sub>2</sub> capture and storage (CCS).

According to the Intergovernmental Panel on Climate Change (IPCC) 5th Assessment Synthesis Report (2014), without CO<sub>2</sub> sequestration, it may not be possible to meet the emission reduction target of limiting global warming to below 2 °C relative to pre-industrial levels. In addition, the panel concluded that without CO<sub>2</sub> sequestration, the cost of mitigation will rise by an average of 138%, which is more than double the cost. In order to maximise the storage capacity, researchers and scientists have considered various types of geological formations to store and isolate captured anthropogenic CO<sub>2</sub> from the surface environment. Three major potential geological formations have recently attracted attention; 1) depleted oil/gas reservoirs, 2) un-mineable coal beds, and 3) deep saline aquifers. CO<sub>2</sub> sequestration in depleted oil and gas reservoirs not only provides a solution to mitigate global warming,

it also has economic value which can be used to recover the high cost of carbon capture and storage (CCS). Although this method is considered as the most mature technology for injection of CO<sub>2</sub> for enhanced oil recovery (EOR), the limited available capacity, potential leakage risks and high production costs involved in the process, may limit its feasibility as a CO<sub>2</sub> storage site. Un-mineable coal beds can be used to permanently store injected CO<sub>2</sub> due to the adsorption capability of CO<sub>2</sub> to the coal matrix. The ability to offset the operational costs by using the valuable energy product methane (CH<sub>4</sub>) is a unique advantage associated with this process. However, a comprehensive review of CO<sub>2</sub> sequestration in deep un-mineable coal beds with the recovery of methane gas by White et al. [34] highlighted several key issues associated with this process, including limited global storage capacity, storage integrity, physical and chemical processes, and environmental health and safety. Moreover, Stenvens et al. [32] revealed that many of the potential coal seams have relatively low permeability, causing this process to not be feasible as a CO<sub>2</sub> storage candidate.

CO<sub>2</sub> geo-sequestration in deep saline aquifers has recently attracted the attention of many researchers, due to its many unique advantages [34]. Deep saline aquifers are the most abundant geological reservoirs in the sub-surface and can be found both on- and off-shore [5]. According to a comprehensive review of several concurrent projects by White et al. [34]; CO<sub>2</sub> sequestration in deep saline aquifers is the most technically feasible approach with few or no negative environmental impacts. In addition to these advantages, deep saline aquifers have the largest storage capacity of all geological media [5], and it is estimated that up to 10,000 GtCO<sub>2</sub> could be stored worldwide in aquifers.

However, sequestration of this ever-increasing anthropogenic CO<sub>2</sub> needs suitable deep underground reservoir formations with huge CO<sub>2</sub> storage capability and safety [5]. To satisfy these two requirements, a host formation with adequate strength, permeability and porosity is required [29]. The adequate formation permeability and porosity facilitate relatively fast movement of injected CO<sub>2</sub> into tiny pores which ensures the safety of the storage process, allowing sufficient time for hydrodynamic trapping to occur within a short timescale. The formation strength is also important to avoid premature failure of the well and to avoid cross-formational leakage or even direct release of CO<sub>2</sub> to the surface [25]. In addition, carbonate- and silicate-cemented sandstone formations have been identified as potential reservoir rock formations [22,26] because they provide a more permanent sink for CO<sub>2</sub> by long-term mineral trapping in the form of silicate and carbonate minerals. It is therefore important to have comprehensive knowledge of the CO<sub>2</sub> storage process in these particular types of geological reservoir rock, which requires the precise identification

**Table 1**  
Amount of CO<sub>2</sub> emitted from main anthropogenic sources in 2011 [12,13].

Anthropogenic sources of CO <sub>2</sub>	% of CO <sub>2</sub> emission	Remarks
1) Fossil fuel use	87%	Coal is responsible for 43% of carbon dioxide emissions from fuel combustion, 36% is produced by oil and 20% from natural gas.
Electricity and heat generation sector	41%	
Industrial sector	20%	
Transportation sector	22%	
Road transport	72%	
Marine shipping	14%	
Global aviation	11%	International flights create about 62% of these emissions with domestic flights representing the remaining 38%. contributed 3.3 billion tonnes of carbon dioxide emissions in 2011
2) Land use changes	9%	
3) Industrial processes	4%	Making 1000 kg of cement produces nearly 900 kg of carbon dioxide 1.9 tonnes of CO <sub>2</sub> are emitted for every tonne of steel produced
Cement production		
Steel production		

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